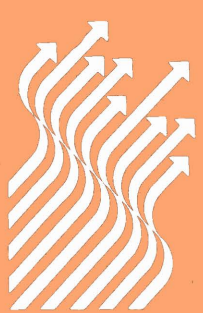
Commission of the European Communities

environment and quality of life

The air quality standard for SO_2 and particles (Directive 80/779/EEC) and its significance for the other main air pollutants



Report

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The air quality standard for SO_2 and particles (Directive 80/779/EEC) and its significance for the other main air pollutants

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I

EXECUTIVE SUMMARY

1. In March 1985 a study group of the Commission published a report on the necessity of a long-term limit value for NO_2.[1] In this report, the experts stressed the importance of combined effects of air pollutants. Inspired by the report, the Commission launched a follow-up study with the aim of

   i. compiling information on limit and guide values set out in industrialized countries and to identify those which are actually combined ones;

   ii. reviewing the relevant literature on combined effects of the pollutants NO_2, SO_2 and O_3 and particulate matter on human health and the environment;

   iii. deriving, if possible, non-adverse effect levels for combinations of these pollutants;

   iv. comparing these combined non-adverse effect levels with values derived for the effects of single pollutants;

   v. comparing the combined non-adverse effect levels with pollutant concentrations measured in ambient air;

   vi. advising the Commission in which way the findings of this study, if necessary, could be incorporated into the actual air pollution abatement policy.

[1] COM(85) 371

2. With regard to point i., the authors identified only very few cases in which combined limit or guide values are set out in industrialized countries; examples are the SO_2- and black smoke limit values of Directive 80/779/EEC and some ambient air limit values for SO_2 and NO_2 and SO_2, No_2 and Formaldehyd, respetively, implemented in the USSR.

   Combinations of pollutant concentrations are more common in national alert systems, where the emergency levels of two or more pollutants are linked to each other. However, these combinations are usually not defined to take better account of combined effects but for air quality management purposes.

3. Theoretical considerations of the question in which way pollutant limits could be linked to achieve combined values resulted in the recommendation of two possible approaches, which have to be selected according to the background of a particular case:

   - the laying-down of a stringent value for a single compound only so that even if an attack of this compound in combinations with other compounds occurs adverse effects cannot happen (worst case approach);

   - the index approach in which the limit value is built up from additive elements, e.g.

$$\text{Index} = \frac{C^1}{LV_1} + \frac{C^2}{LV_2} + \ldots$$

   where C_1 and C_2 are the measured concentrations and LV_1 and LV_2 are the limit values for the compounds 1 and 2.

4. From the comprehensive review of literature on combined effects it could be concluded that:

   - controlled human exposure studies have not consistently demonstrated any enhancement of respiratory effects for combined exposures of O_3, NO_2 and CO; however there is no doubt that the interaction of SO_2 and particulate matter is an important synergistic factor;

   - the present understanding of how pollutant combinations influence plant growth and development is still fragmentary. However, data have demonstrated that interactions can occur if combinations of O_3 and SO_2, SO_2 and NO_2 and SO_2, NO_2 and O_3 attack simultaneously.

   - the review of combined attack on materials gave no sound basis for any working hypothesis suitable for this study.

5. Based on the identified effects on human health and plants the following preliminary non-adverse effect levels could be derived.

   For the protection of human health:

	Concentration in ug/m^3	
	SO_2	Black smoke
24 h mean	100--150	100--150
Annual mean	40--60	40--60

   For the protection of the environment:

	Concentration in ug/m^3		
	SO_2	NO_2	
24 h mean	150	300	
Annual mean	40	40	
and			
24 h mean	SO_2	NO_2	O_3
Annual mean	60	30	30

These are values for pollutants as they occur in the gasphase of ambient air. It should be underlined that effects due to acid deposition were not subject of the study.

6. Current non-adverse effect levels for single compounds are considered to lay in the following ranges:

Compound	Non-adverse effect levels in ug/m^3 for the protection of Human health	Vegetation
SO_2	) Little data on single effects available, but effects due to the impact of single compounds are less important than combined effects	50--120, as annual mean
Black smoke		
NO_2	120--300, as 1 h max. level	4000--9000, as 1/2h value 250--300, as 7 month average
O_3	120, as 1 h max. level	3--1000, as 1/2h value 50--300, as 4 h average

7. The comparison with the combined non-adverse effect levels mentioned above shows that:

with regard to SO_2: the single non-adverse effect levels for SO_2 alone for the protection of human health and the environment are more or less equal to the levels of combined effects as far as the annual mean levels are concerned, but less stringent for the short-term effects.

with regard to NO_2: the single short-term non-adverse effect level for the protection of human health is more stringent than the one for combined effects on vegetation, while in the case of the long-term non-adverse effect levels the one for combined effects on vegetation is much more stringent than the one for NO_2-effects alone.

with regard to O_3: the non-adverse effect levels for combined effects are not comparable to those of O_3 alone, because effects due to the combined impact of air pollutants happen during long-term periods while those for O_3 alone within short-term periods.

Generally it can be said that the uncombined limit values laid down in industrialized countries are less stringent than the values established as non-adverse effect levels in this report and outlined in point 5.

However, even if the single non-adverse effect values are higher, they may be more stringent than the lower combined values because exceeding the two lower limit values at the same time may be less likely than exceeding one higher single value. All depends on the actually chosen value and the local situation. The final answer about the stringency of one or the other set of limit values can in most cases be answered only by a comparison of measured data.

8. Investigations in the actual pollutant levels, as measured at different site categories in the EC Member States showed the following concentration ranges in ug/m^3:

		SO_2	NO_2	O_3	Black smoke
urban sites	annual mean	40--160	20--100	20--40	5--80
	24 h mean	40--350	40--300	60--300	5--350
rural sites	annual mean	5--30	5--30	50--80	1--9
	24 h mean	10--150	20--60	60--300	3--36

The detailed comparison of these values, taking into account combined time patterns of two or more pollutants at one site, showed that:

- the combined non-adverse effect levels for SO_2 and black smoke are exceeded in several urban areas, but not in rural areas;

- the combined non-adverse effect levels for NO_2 and SO_2 are most likely exceeded in some urban areas, but not in rural areas;

- the combined non-adverse effect levels for SO_2, NO_2 and O_3 are most likely exceeded in several urban areas, but not in rural areas.

However, one should bear in mind that:

i. these non-adverse effect values had to be derived partly from incomplete information;

ii. such low limit values will probably, for political reasons, not be accepted by Member States in the near future;

iii. exceeding the non-adverse effect values in urban areas will not automatically result in any damage because a large number of sensitive plants cannot be found at all in such areas.

Therefore, one should consider the findings mentioned in this point much more as a preliminary indicator of still existing problems rather than as a basis for the formulation of a detailed abatement strategy.

9. In a separate subchapter, some basic reflections on possible air quality management systems (AQMS) were made. It could be demonstrated that the classical AQMS which is based on ambient air quality standards and where the quality of the air is managed to meet these standards cannot generally be applied anymore. In particular the "imission standard" approach, as outlined in all its principles in chapter IV.2 cannot be applied anymore nationwide because:

   - clear "non-adverse effect levels" which guarantee the protection of human health and, in particular, the environment are difficult to define;
   - even if they could be defined it would be very difficult to identify the single sources responsible for a certain pollution in a particular rural area;
   - adequate monitoring, a necessary prerequisite for any "immission standard" approach, is difficult to perform in rural areas.

   The only appropriate policy to therefore:

   i. to reduce the relevant emissions in urban areas down to those levels identified as non-adverse effect levels;

   ii. to introduce a long-term policy of an overall reduction of emissions from all sources important for long-range transport of pollutants.

Therefore it is proposed to implement a comprehensive emission standard strategy that establishes permitted levels of emission for specific groups of emission sources. Permitted levels can be applied to any category of emitters, generally by processes and/or any variety of geographical areas. Often they can be associated with air quality standards in urban and industrial areas. However, for pollution in rural areas by long-range transport they have to be set out as an entirely independent strategy that constitutes a form of the "best practical means approach". An important element to decrease emissions in applying such a strategy is the permanent improvement of the state of-the-art of abatement technologies and the development of new industrial procedures with low pollution.

A supplementary element for some cases may be the laying down of product norms.

10. With regard to the SO_2-abatement-policy, the authors concluded that the Commission, by attacking emissions from large power plants and large industrial combustion installations by its proposal COM(83) 704 and emissions from households by its proposal COM(85) 377, is already on the right road towards cleaner air.

However, both proposals have some shortcomings:

i. the proposal on large combustion installations does not include any financial support system and/or derogations for "poor" Member States and is therefore unacceptable for some Member States;

ii. the proposal on the S-content of gasoil lays down a lower limit value which is unnecessary and which makes local policies more difficult.

In total, these approaches would reduce total SO_2-emissions, if adopted, by approximately 37%, equal to 5800 kilotonnes. Supplementary measures are possible and necessary:

i. the setting out of limit values for industrial sources;
ii. the limiting of the S-content in coal and heavy oils.

With regard to point i. emission standards should be laid down for all combustion installations larger than 5 MW therm. The standards should be in such a range that washed coal (0.8% - 1%S) and low sulphur oil (1%-1,2%S) is allowed to be burnt and no further post-treatment of the exhaust gases is needed in order to comply with the limit values.

With regard to point ii. it should be forbidden to burn oil with more than 0,3% S and coal with more than 0.8% S in domestic installations. Moreover the Commission should enforce its activities on energy savings and research and development work on clean technologies for industry.

It has been estimated that such measures would cut down SO_2-emissions by 40% from industrial sources and by 30% from domestic sources, resulting in a supplementary reduction of approximately 2000 kilotonnes, equal to 13% of actual SO_2 emissions. Together with the already proposed measures an overall reduction of 50% would be achieved. Finally, it is absolutely necessary to accompany these measures by political action on the ECE-level because the Member States of the EC are only responsible for approximately 25% of total European SO_2 emissions. Any further reductions in the EC Member States would be small in comparison with the emission of others, in particular East European countries.

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11. With regard to the NO_x-reduction policy, the Commission has submitted to Member States two main proposals with the aim to reduce emissions from two sources:

- emissions from passenger cars will, most likely, be reduced due to the Council agreement of June 1985, based in the Commission's proposal COM(85) 288;
- emissions from large combustion installations are attacked by proposal COM(83) 704, which aims at a 40% overall reduction of emissions from this source.

The implementation of these proposals would result in an overall reduction of NO_x-emissions of approximately 3000 kilotonnes by 1995, equal to 29% of total man-made emissions.

As in the case of SO_2, supplementary measures are possible, which may result in further substantial reductions:

i. the laying down of limit values for motor vehicles above 3.5 tonnes weight;

ii. the laying down of emission standards for combustion installations in the range of 4 MW therm and 50 MW therm, including industrial boilers.

Such measures would reduce NO_x-emission by approximately another 900 kilotonnes, equal to 9% of total man-made emissions, so that an overall reduction of 38% seems possible.

Measures, as proposed in point i. are underway and the Commission will put forward an appropriate proposal by the end of 1985.

Proposals concerning point ii. are under consideration and being studied by working groups of the Commission.

As in the case of SO_2, it is recommended to enforce activities on ECE-level because a substantial part of total man-made NO_x is emitted in non-Member States of the EC. An efficient European overall reduction strategy requires that all European countries reduce emissions.

12. As far as particulates are concerned, two sources are of major importance:
    - industries
    - power plants.

With regard to power plants, a proposal has already been made by the Commission (COM(83) 704), which would, if adopted, result in a 40% reduction of emissions from this source, equal to approximately 9% reduction of overall emissions.

The main source of particulate emissions, industries, is however still not covered by the Commission's activities and it is recommended that appropriate proposals should be tabled. It is extremely difficult, due to the lack of data, to estimate the reduction potential of such proposals because emissions have already been substantially reduced in the last decade by some Member States. Others, however, have done little. However, a 30% reduction of emissions from industrial sources seems to be still possible on a EC-wide level, resulting in a 17% reduction of overall emissions. As in the case of SO_2 and NO_x, it seems that the best way of achieving such reduction is by setting out emission standards for the various industries. Old installations should be obliged to meet the standard with an appropriate delay.

13. With regard to photochemical oxidants (O_3) it is still too early at this stage to give any generally valid figures for the reduction in oxidant levels which could be expected from any given measure to limit emissions of individual categories of precursor. To provide a solid basis for decisions to reduce oxidant levels, first physical and chemical dispersion models must be further developed. Regional measures to reduce emissions are unlikely to be enough in themselves to keep ozone levels within non-effect limits. On the other hand, regional measures could probably prevent regional or local peaks. Consequently, a coordinated campaign to reduce emissions throughout the whole of Europe appears the only way to reduce the ozone load over wide areas.

    Nevertheless, it is fair to assume that any kind of reduction in precursor emissions will reduce the average total pollution load over wide areas, even if it fails to produce a significant reduction in the ozone load locally.

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Introduction and General Aspects

Very often it is said that ambient air is the largest chemical laboratory of the world. This is most likely true: thousands of different compounds are dispersed in ambient air, interacting with each other, creating new compounds, getting solved in water droplets, getting absorbed by surfaces and so on.

All more or less stable products of such processes can have an impact on human beings, animals, terrestrial and aquatic ecosystems and materials.

Although our present knowledge of such impact is still very incomplete, in a couple of cases dose-response relationships between mixtures of air pollutants and a particular place of impact have been established.

The best studied cases are the combined effects of sulphur dioxide and particulate matter on human health and the combined effects of sulphur dioxide, nitrogen oxides and ozone on vegetation.

However, due to the large number of parameters involved in such studies the number of experiments necessary to get a complete picture of the impact is huge. Therefore, even in the best studied cases, our understanding is still incomplete and very often guesswork. Inspite of these uncertainties concepts of an appropriate air quality management have to be developed in order to protect human health and the environment. The development of such concepts is already difficult for only one single pollutant because, apart from the uncertainties on the effect side, answers to complex questions have to be found: e.g. economic consideration like cost/benefit studies, control problems like extent of necessary survey and so on. However, the need of such studies is evident even in the light of the already existing but limited knowledge of combined effects because control strategies which do not take into account the real atmospheric conditions but are limited on single compounds, might be extremely uneffective.

The aim of this study is not to find an answer to all questions involved in this complex business. The goal is to discuss certain aspects of an appropriate control strategy for particular cases and to contribute in this way to the on going research for the best practical air quality management.

For the purpose of this study it is important to develop terminology that distinguishes between the different categories of response to the effects of two or more pollutants.

This will be done as follows for the case of two pollutants.

When one pollutant (A) causes no effect but the second (B) does, it is termed "single effect". This study, however, does not deal with such cases. The concept of a "combined effect" implies that both pollutants have some effect on the one and the same subject. This concept has to be further divided into subcategories:

1. Additive response

   Effect (AB) = Effect A + Effect B

2. Interactive response

   Effect (AB) $\neq$ Effect A + Effect B

The interactive response may be of two possible types:

2.a. Synergism

   Effect (AB) $>$ Effect A + Effect B

2.b. Antagonism

   Effect (AB) $<$ Effect A + Effect B

Two examples may help to illustrate these categories:

- An additive response is the corrosion of materials due to the combined attack of SO_2 and NO_2. Both gases can form acid substances and increase the corrosion rate. Therefore the combined effect "corrosion" is the sum of the single effects of corrosion due to SO_2 and NO_2.

- An interactive synergistic response is the combined effect of SO_2 and suspended particulate matter (SPM) on human health. Both pollutants effect the human respiratory system in different ways and cause a response which is different from the response caused by only one of them. This qualitatively new response to the combined action of SO_2 and SPM has been identified at lower concentration levels than the responses due to the action of SO_2 or SPM as single pollutants.

For the purposes of this study the interactive synergistic effect is the most important one because it requires, in the most cases, countermeasures at comparatively low concentration levels.

Additive responses are also of interest. However, they involve the whole question of "acid rain" which is studied intensively by other EC-working groups. Therefore this category of combined effects will be not dealt with in this study.

I. REVIEW OF AMBIENT AIR QUALITY STANDARDS AND EMERGENCY LEVELS IMPLEMENTED IN INDUSTRIALISED COUNTRIES

I.1. Introduction

In this chapter a short review of ambient air standards and emergency levels implemented in industrialised countries is given. The aim of this review is to identify cases where combined effects are the raison for setting such values.

Ambient air quality standards define - on the basis of current knowledge on dose-effect-relationships - the level of pollutants in ambient air which is considered to be "acceptable" in terms of concentrations or deposition fluxes of the respective pollutants.

They serve as management targets, defined in quantitative terms, guiding the development of abatement strategy plans and they permit the checking of a plan's effectiveness.

These targets are termed "standards" if they are specified by law, decree or any other enforceable regulation; otherwise they are termed "objectives" or "goals".

The basis for setting a standard is the scientifically established relationship between given concentrations of one or more pollutants in ambient air on one hand and the related effects on the other. These relationships are termed "air quality criteria".

The effects may be effects on human health, animals, vegetation, soil, aquatic ecosystems, materials, visibility or climate. Accordingly, different standards may be applied. There may be acute effects which cause an abrupt change in the condition of the receptor; there can also be chronic effects where an impairment in the condition of the receptor can occur due to exposure over a long period of time.

Accordingly, a standard is defined normally in a twofold way: as a long-term standard, e.g. as annual average or as the median of a frequency distribution; and as a short-term standard, e.g. as a maximum value or - preferably - as a high percentile of a frequency distribution.

It is essential to keep in mind these statistical implications in a standard's definition when comparing standards. It has also to be kept in mind that the aforementioned criteria - though they are providing the scientific basis for setting a standard - are only one element within the decision process: The actual setting of a standard is a political decision whereby the degree as to which society is willing to tolerate adverse effects is evaluated, taking into account economic, technical, political and other aspects. Due to the fact that the relative importance of these aspects may differ from country to country and due to our incomplete knowledge concerning dose-effect-relationships, it is not surprising that different ambient air quality standards for a given substance have evolved in different countries.

There is another important aspect which has to be considered when setting standards or when evaluating standards applied in different countries. This aspect refers to the assessment system. The standard on one hand and the assessment system on the other form the two sides of the coin. The assessment system provides the statistics on ambient pollutant situation which have to be compared with the standard in order to check whether or not the ambient situation is in compliance with the standard. But, as our picture of reality depends strongly on the way we look at reality, it is of vital importance to specify the assessment system as precisely as possible. This implies that the standard and the assessment system are both strongly interrelated.

The assessment system for a given substance consists of several building blocks:

- number and location of monitoring stations
- sampling time/sampling frequency
- instrumentation
- calibration
- data evaluation/data reduction
- modeling techniques.

All these building blocks and their associated elements comprise what has been defined "the standards". And if any one of these blocks is changed, a resultant change also occurs in the definition of the standard, even if it remains numerically constant.

I.2. Examples of ambient air quality standards

The review of ambient air standards set out in a number of industrialised countries shows that in general no combined standards for SO_2, NO_2, HC, O_3 and particular matter are laid down (see tables I.2.1 to I.2.6).

There are two exceptions to be mentioned. Firstly, the Directive 80/779/EEC, which combines the SO_2 limit values with "trigger values" for black smoke (see table I.2.1) and secondly the combination of several limit values in the USSR (see table I.2.1).

The approach of the European Commission which has been implemented by seven of the ten Member States, is based on health considerations of combined effects. However, the values laid down in the Directive are considerably higher than those recommended by the WHO.

The ambient air quality standards of the USSR are calculated by a scientific committee and approved by the Ministry of Public Health. This approval gives a semi-legal value to these standards. These are valid for urban areas. Most of the standards laid down in Russia are standards for single compounds; however, in a number of cases combinations with other compounds have to be taken into account.

Two cases are of interest in this study:

1) $SO_2 + NO_2$

Short-term limit value:

$$\frac{C_{NO_2}}{85 \text{ ug } NO_2/m^3} + \frac{C_{2O_2}}{500 \text{ ug } SO_2/m^3} \leqq 1$$

Long-term limit value:

$$\frac{C_{NO_2}}{85 \text{ ug } NO_2/m^3} + \frac{C_{SO_2}}{50 \text{ ug } SO_2/m^3} \leqq 1$$

2) $O_3 + NO_2$ + Formaldehyd

Short-term limit value:

$$\frac{C_{O_3}}{LV_{O3}} + \frac{C_{NO_2}}{85 \text{ ug } NO_2/m^3} + \frac{C_{CHO}}{12 \text{ ug CHO}/m^3} \leqq 1$$

Long-term limit value:

$$\frac{C_{O_3}}{LV_{O3}} + \frac{C_{NO2}}{85 \text{ ug } NO_2/m^3} + \frac{C_{CHO}}{12 \text{ ug CHO}/m^3} \leqq 1$$

No general limit value for O_3 has as yet been laid down. It is foreseen that in such cases appropriate values should be formulated for each situation.

The short-term value is based on a 24 h average. The long-term value is an annual average.

I.3. Examples of emergency levels

The review shows that in most cases values for single compounds are only laid down as alert concentrations (see table I.3.1). However, in few cases combined values have been defined:

- in the F.R.G. SO_2 + SPM will be combined in future regulations by the formula:

  $$SO_2 + 2 * SPM$$

- in Spain SO_2 + SPM are combined by the formula:

  $$SO_2 * SPM$$

- in California O_3 and SO_2 are combined by the formula:

  $$O_3 + SO_2, \text{ if } O_3 \leqq 0.1 \text{ ppm and } SO_2 \leqq 0.1 \text{ ppm}$$

The concentration levels laid down in these regulations are, of course, much higher than those defined as air quality standards. However, the study shows interesting examples of the way how such combined values can be linked to each other.

I.4. Considerations on appropriate definitions of combined limit values

The given short review of air quality and emergency levels showed a number of different possibilities for the laying down of limit values based on combined effects. In principle it can be divided into the following approaches:

1. The definition of an index which should not be exceeded and which is built up from additive elements, e.g.

   $$\text{Index} = \frac{C_1}{LV_1} + \frac{C_2}{LV_2}$$

   where C_1, C_2 are the measured concentrations and LV_1, LV_2 are the limit values for compounds 1 and 2.

2. The definition of a single limit value which is built up from added concentrations, e.g.:

$$LV = C_1 + C_2 + \ldots$$

3. The laying down of a single limit value combined with trigger values (TV) of other compounds, e.g.

$$LV_{A1} = C_{A1}, \text{ if } TV_1 \leqq C_{B1}$$

$$LV_{A2} = C_{A2}, \text{ if } TV_2 \leqq C_{B2}$$

with LV_{A1} = limit value of compound A

C_{A1} = concentration of compound A

TV_1 = trigger value

C_{B2} = concentration of compound B (trigger compound)

4. A limit value which is based on multiplication of concentrations, e.g.

$$LV = C_1 * C_2$$

5. The definition of a single value which already takes into account the worst case of possible combined effects without requiring any combined survey or control, e.g.

LV = most stringent value identified for single and combined effects.

The question which of these possibilities is the most appropriate to be chosen cannot be answered generally. In principle each case has to be investigated separately.

However, cases 3 and 4 seem to have basic practical disadvantages. Case 3 includes a strong dependence of the limit value on the trigger value. This can be demonstrated with the aid of the combined values as laid down in Directive 80/779/EEC for SO_2, triggered by black smoke. At two stations in Doncaster (United Kingdom), the measured black smoke concentration was, in the period 83/84, just equal to 40 ug/m^3 so that

the higher SO_2 limit value of 120 ug/m^3 had to be applied. However, the measured SO_2 concentrations were not above 120 ug/m^3 but above 80 ug/m^3. If the BS-value had been 41 ug/m^3 that means only 1 ug/m^3 more, the limit value of 80 would have to be applied and a violation of the Directive would have occurred. It is known that the measurement of BS is of limited precision, e.g. in the range of ± 15%. Consequently the uncertainty of black smoke measurements can decide whether one or the other SO_2 limit value has to be applied.

Case 4 is, due to the multiplication rule, too sensitive to small changes in one or the other value, e.g. if the limit value is equal 1000 and C_1 is equal 33 all can depend on whether C_2 is equal to 30 or to 31. Again the precision of actual measurement is unsufficient for the application of such an approach because ambient air measurements are, in most cases, not reliable to within ± 1 unit.

If the first priority is the protection of the environment, the best approach is the definition of the most stringent single limit value for a certain compound taking into account combined effects (case 5). Moreover, it is easier to control compliance and therefore implies the maximum margin of safety for all measurement problems. However, the laying down of such a stringent value may not be cost-effective.

Approaches 1 and 2 are quite similar. They are implemented in some countries and can be considered as a compromise between the stringent approach 5 and the less reliable approach 3.

I.5. Conclusions to Chapter I

1. The review of ambient air quality standards and emergency levels showed that, only in very few cases have combined levels been laid down.

2. Where such an approach was chosen, different ways of combining one concentration with others were defined and these can be divided into three main cases:
   i) addition of the concentrations,
   ii) multiplication of the concentrations,
   iii) change of the limit value of one compound if a certain value of the second compound is exceeded (trigger approach).

3. A fourth possibility is to set out a low single value so that even if the concentrations of other compounds exceed certain values adverse effects cannot happen. Such an approach was probably chosen by Switzerland. In such cases no combined values have to be defined explicitly because the adverse combined effects can be avoided by one single value.

II. Examples of combined effects of air pollutants

The content of this chapter is a limited review of combined effects of air pollutants on human health, ecosystems and materials. The aim of this review is to identify areas where combined effects may require more stringent counter measure than are necessary to counteract single effects.

It should be underlined that the authors do not feel competent to give recommendations for limit values for the protection of human health and/or the environment. However, the definition of appropriate air quality management strategies requires boundary conditions coming from the effect side. Therefore, this review provides data which lead to the definition of a realistic working hypothesis for the study of strategies.

For practical reasons the review deals only with combined effects of sulphur dioxide, nitrogen oxides, ozone and suspended particulate matter (SPM). Other substances like heavy metals and hydrocarbons will be taken only into consideration if absolutely necessary.

The following sub-chapters are based on comprehensive studies carried out and published by national and international bodies like the World Health Organisation, the US Environmental Protection Agency, the German Umweltbundesamt and others.

II.1. Combined effects on human health

Although human beings, animals and the environment are always exposed to a complex mixture of gases and particulates, comparatively few studies on combined effects of NO_2, SO_2, O_3 and SPM have been conducted in the past.

However, in most cases air pollutants effect human health as single compounds and only in a few cases, e.g. due to the formation of metabolic substances in the body, can synergistic responses be expected. As it will be demonstrated the interaction of SO_2 and SPM is an exception to this rule.

Besides controlled exposure studies on animals and human beings some epidemiological studies have been carried out. Due to the difficulties in relating results from animals studies to the response of human beings the following review does not take them into account but focuses on controlled exposures and epidemiological studies.

II.1.1. Combined effects of SO_2 and O_3

Sulfur dioxide and ozone (O_3) may combine to form sulfuric acid on the warm, moist surfaces of the respiratory tract. Studies have not yet demonstrated, however, that a true synergistic bond exists between SO_2 and O_3.

BATES and HAZUCHA (1973) and HAZUCHA and BATES (1975) exposed eight volunteer male subjects to mixture of 700 ug/m^3 O_3 and 900 ug/m^3 SO_2 for 2 hours. Sulfur dioxide alone had no detectable effect on lung function, while exposure to ozone alone resulted in decrements in pulmonary function. The combination of gases resulted in more severe (10 to 20 % decrement) respiratory symptoms and pulmonary function changes than did ozone alone. Moreover, the effects resulting from O_3 and SO_2 in combination were apparent in within 0.5 hours, in contrast to a 2-hour time lag for exposure to O_3 alone.

BELL ET AL. (1977) attempted to replicate these studies with only four normal and four ozone-sensitive subjects. They showed that an O_3 + SO_2 mixture had greater detrimental effect on all pulmonary functions measured than did O_3 alone. However, only some of these parameters showed statistically significant decrement when compared to O_3.

None of the subjects responded in a similar manner in the Bell study. Retrospective sampling of the ambient air conditions utilizing particle samplers and chemical analysis in the chamber showed that acid sulfate particles could have been 10- to 100-fold higher in Hazucha and Bates' chamber and thus might have been responsible for the synergistic effects observed.

HORVATH, FOLINSBEE (1977) and BEDI (1979) exposed nine young men (18 to 27 years old) to 800 ug/m^3 O_3 and 1060 ug/m^3 SO_2 singly and in combination for 2 hours in an inhalation chamber at 25°C and 45 percent RH. Subjects exposed to filtered air or to SO_2 showed no significant changes in pulmonary function. When exposed to either O_3 or O_3 plus SO_2, the subjects showed significant decreases in maximum expiratory flow, forced vital capacity, and inspiratory capacity. However, there were no significant differences between the effects of O_3 + SO_2 and O_3 alone, thus, no synergistic effects were discernible in their subjects.

KAGAWA and TSURU (1979) conducted chamber studies with six subjects. They found a significant decrease in five of the six subjects (5/6) exposed to O_3 alone. In three of the six young male subjects, they found a significant enhanced decrease in specific airway conductance (SG_{aw}) after exposure to the combination pollutants compared to the decrease in SG_{aw} in these subjects to O_3 exposure. Two other subjects had similar decreases to either O_3 or O_3-SO_2 exposure. They further suggest that the effect of the two gases on SG_{aw} is more than simply additive and results from a combined pollutant exposure. Subjective symptoms of cough and bronchial irritation were reported to occur in subjects exposed to both O_3 and O_3-SO_2.

Epidemiological studies by KAGAWA ET AL (1975, 1976, 1983) give rise to concern that O_3 associated with SO_2 increase airway resistance. However, even in the light of the current knowledge, the question of potential synergistic interaction between SO_2 and O_3 remains unresolved.

II.1.2. Combined effects of O_3 and NO_2

HORVATH and FOLINSBEE (1979) exposed eight young adults to either filtered air or 980 ug/m^3 O_3 plus 940 ug/m^3 NO_2 in filtered air under different environmental conditions. The pulmonary responses to ozone alone were as found in previous studies by the same research group. No additive effect or interaction between ozone and nitrogen dioxide was observed. LINN ET AL. (1976) also came to the same conclusions in an extensive epidemiological study.

II.1.3. Combined effects of NO_2 and suspended particulate matter

NAKAMURA (1964) studied the interaction of NO_2 and sodium chloride aerosol and found that sodium chloride had no influence on airway resistance R_{aw}. When the sodium chloride aerosol (mean diameter 0.95 um) was added to the exposure atmospheres, the increases in R_{aw} for the group were approximately 40 percent, about twice that produced by the gas alone. A sodium chloride aerosol comprised of smaller particles (mean diameter 0.22 um) at 1,400 ug/m^3, in combination with the same concentrations of NO_2, produced no increase in R_{aw} over that caused by the gas alone. The consistent sequential methodology used in this study tends to reduce the credibility of this study; nevertheless, the fact that the final exposure challenge in the sequence increased the R_{aw} when the NaCl aerosol particles averaged 95 um in diameter but did not increase R_{aw} when they averaged 22 um in diameter indicates that when used in the same sequence the larger particles of NaCl enhanced the effect of NO_2 while the smaller particles did not.

II.1.4. Combined effects of O_3 and suspended particulate matter

Possible interactions between O_3 and SPM have been reported in connection with decreased expiratory flow in children (LEBOWITZ ET AL. 1982, 1983, LEBOWITZ 1984) and adults with symptoms of airway obstructive disease. However, available data do not allow final conclusions at present.

II.1.5. Combined effects of SO_2 and NO_2

In exposure chamber experiments with 11 subjects VAN NIEDING ET AL. (1979) detected increased air resistance. However, the exposure combinations (15 ppm of each gas) were unrealistically high. LINN ET AL. (1980) could not find any changes in pulmonary functions when exposing healthy persons and asthmatics to more realistic concentrations (0.5 ppm of each gas).

SPEIZER and FERRIS (1973) and BURGER ET AL. (1973) could not detect any difference in various pulmonary function tests when examining 128 traffic policemen in Boston urban area and compared results to pulmonary functions of 140 patrol officers in suburban areas.

II.1.6. Combined effects of SO_2 and suspended particulate matter

The combined action of SO_2 and SPM on human health can be considered as the best documented synergistic effect of air pollutants.

From the large number of publications only the most important are briefly reviewed in the following pages.

Suspended particulate matter has been shown to be potentially important in enhancing the effects of SO_2 exposure. Airway resistance increased more after combined exposure to SO_2 and sodium chloride in several studies, although others have failed to reach the same conclusions (see Table II.1.-1). $MEF_{50\%}$ (maximal expiratory flow rate at 50 % vital capacity) was found to be significantly reduced after exposure to a combination of saline aerosol and 13,100 ug/m^3 SO_2 (SNELL and LUCHSINGER, 1969). KOENIG ET AL. (1980, 1981) studied pulmonary function changes and observed in extrinsic asthmatics both at rest (1980) and during exercise (1981) with exposure to 2,620 ug/m^3 SO_2 and 1 ug/m^3 NaCl. Significant decreases in $\dot{V}_{max50\%}$ and $\dot{V}_{max50\%}$ were observed under aerosol conditions both at rest and during exercise for asthmatics but not for all normals. However, NaCl alone did not produce such effects, suggesting that either the exercise or the combination of SO_2 and NaCl was important.

The most important evidence of the synergistic effect of SO_2 has been derived from epidemiological studies (see Table II.1.-2).

In general, the epidemiological studies provide evidence for severe health effects, such as mortality and respiratory disease being associated with marked elevations of atmospheric levels of SO_2 and Black Smoke (BS).

MARTIN and BRADLEY (1960) and MARTIN (1964) dealt with a relatively small body of data from London in the late 1950s. No clear "threshold" levels were revealed by their analyses regarding SO_2 or BS levels at which significantly increased mortality began to occur. Based on their findings, there appears to be little question that mortality in the elderly and chronically ill was elevated in association with exposure to SO_2 and BS levels in the range of 500 - 1000 ug/m^3. Greatest certainty applies for levels in excess of 700 - 750 ug/m^3.

LAWTHER's morbidity studies (1958, 1970, 1975) suggest that acute exposure to 24 hrs of pollution levels of 200 - 500 ug/m^3 BS in association with 500 to 600 ug/m^3 SO_2 are likely to cause a worsening in respiratory disease symptoms in chronically ill bronchitis patients. The LAMBERT and REID study suggests that respiratory disease symptoms (cough and phlegm) are associated with long-term exposures of adults to BS levels in the range of 100 - 200 ug/m^3 or above in association with SO_2 levels in the range of 150 - 200 ug/m^3 or above.

Other studies by LINN ET AL. (1967, 1970) suggest that significant respiratory effects occur in children in association with long-term BS-levels in the range of 230 - 300 ug/m^3 together with SO_2-levels of 181 - 275 ug/m^3. However, no effects were seen for children at BS-levels of 48 - 169 ug/m^3 and SO_2 level of 94 - 253 ug/m^3.

SAWICKI (1972) believes to have identified increased respiratory symptoms at the upper limit of this range. No epidemiological data at all are published in the open literature on combined effects of SO_2 and gravimetrically measured particulates. The WHO (1979) in its criteria document 8 only gives reference to unpublished data of SUZUKI and HITOSUGI and concludes that no health guidelines for gravimetrically measured SPM can be given at present.

II.1.7. Combined effects of NO_2, O_3 and CO

HACKNEY ET AL. (1975) exposed four healthy male volunteers to O_3 (1,000 ug/m^3) and subsequently to mixtures of O_3 and NO_2 (560 ug/m^3) or O_3, NO_2 and CO (45,900 ug/m^3). Under these conditions, minimal alterations in pulmonary functions were measured when test subjects were exposed to O_3 alone. These alterations were not increased by the additions of NO_2 or of NO_2 and CO. Another group of seven male volunteers was exposed under a similar protocol with an exposure time of 2 hours and to 500 ug/m^3 O_3. Again, little or no change in pulmonary function was found with O_3 exposure alone, or with addition of NO_2 (560 ug/m^3) or of NO_2 plus CO (45,900 ug/m^3).

Epidemiological studies of HAMMER ET AL. could not find relationships between symptoms like eye irritation, cough, chest discomfort and combined concentrations of CO, O_3 and NO_2 but only to O_3 alone.

II.1.8. Combined effects of SO_2, O_3 and NO_2

VON NIEDING and co-workers (1977) exposed 11 healthy subjects to a combination of NO_2 at 100 ug/m^3, O_3 at 50 ug/m^3, and SO_2 at 300 ug/m^3 for 2 hours, but could not detect an effect on R_{aw} or $AaDO_2$. Exposure to this combination of pollutants did, however, produce what was interpreted by the investigators as dose-depender increases over controls (not exposed to the pollutants) in the sensitivity of the bronchial tree to administered acetylchlorine as measured by increases in R_{aw}. Constriction of the bronchi is a physiological alteration similar to that experienced by many individuals in the form of asthma attacks. The suggestion provide by this study is that exposure to air pollutants may increase susceptibility to asthma attacks in some individuals.

In subsequent experiments, VON NIEDING ET AL. (1979) exposed 11 subjects to O_3, NO_2 and SO_2 singly and in various combinations. The subjects were exposed for 2 hours with 1 hour devoted to exercise which doublec their ventilation. In the actual exposure

experiments, no significant alterations were observed for PaO_2, $PaCO_2$, pH_a and thoracic gas volume (Vtg). Airway resistance total (R_t) and PaO_2 were altered in certain studies. Airway resistance increased significantly in the combination experiments to the same extent as the exposures to NO_2 alone. In the 1-hour post exposure period of the NO_2, SO_2 and O_3 experiment, R_t continued to increase. Subjects were also exposed to 100 ug/m^3 NO_2, 300 ug/m^3, and 350 ug/m^3. No changes in any of the measured parameters were observed.

Studies carried out by ISLAM and ULMER are subject to severe criticism due to a statistically inadequate analysis of data.

However, one can conclude that a combined attack of 280 ug/m^3 NO_2, 140 ug/m^3 O_3 and 810 ug/m^3 SO_2 does not result in any effects (ISLAM, ULMER, 1974).

In epidemiological studies carried out by MAKINO and MIZOGUCHI (1975) symptoms of eye irritation, sore throat, headache and cough were related to oxidant concentration and temperature but not to SO_2, NO_2 or NO.

II.1.9. Combined effects of NO_2, SO_2 and SPM

An epidemiological study of combined effects on pulmonary functions, carried out by MOSTARDI ET AL. (1981) could not find evidence for any adverse effects in the measured concentration ranges (50 ug/m^3 NO_2, 60 ug/m^3 SO_2 and 50 ug/m^3 SPM).

II.2. Combined effects on terrestrial and aquatic ecosystems

Ecosystems represent the natural order by which living organisms are bound to each other and to their environment. They are, therefore, essential to the existence of any species on earth, including man, and as life support systems their value cannot be quantified in economic terms.

Glossery to Chapter II.1.

$AaDO_2$	Difference between alveolar and arterial blood partial pressure of oxygen
DL_{CO}	Difference capacity of the lung for carbon monoxide
FEV_1	Forced expiratory volume (1.0 sec execution time)
FRC	Functional residual capacity
FVC	Forced vital capacity
MEF_{50}	Maximal expiratory flow rate of 50 % vital capacity
MMFR	Maximum mid-expiratory flow rate
$MMFR_{50}$	Maximum mid-expiratory flow rate after 50 % of the FVC has been exhaled
$PaCO_2$	Arterial partial pressure of carbon dioxide
PaO_2	Arterial partial pressure of oxygen
PO_2	Partial pressure of oxygen
R_{aw}	Airway resistance
R_t	Total air resistance
SG_{aw}	Specific airway conductance
$TGV \cdot R_{aw}$	Specific airway resistance, R_{aw} multiplied by the volume at which it was measured
TGV	Thoracic gas volume
$V_{max50\%,75\%}$	Maximum expiratory flow when lung is at 50 % or 75 % of its total vital capacity

Ecosystems are important in the production of food, in the regeneration of essential nutrients as well as atmospheric components, in the assimilation or breakdown of many pollutants from the air, water, and soil, and in energy flow. They also give aesthetic pleasure and improve the quality of life.

Polluting components of ambient atmospheres such as O_3, SO_2, NO_2 and other pollutants may change, modify, or alter ecosystems. The magnitude of these modifications depends on the plant species, cultivar, pollutant concentration, duration and frequency of exposure, and the environmental and edaphic conditions in which plants are grown.

II.2.1. Combined effects of Ozone and sulfur dioxide

More than just an additive reaction on vegetation was noted as early as 1966, concerning Ozone and SO_2. Since then combined effects of O_3 and SO_2 has been extensively studied. Foliage injuries and growth reductions were the most studied effects. Some selected studies on additive ans synergistic foliar-injury and growth responses are summarized in Table II.2.1.-1. Foliar injury symptoms decrease the aesthetic value of various types of woody ornamental and floriculture crop species. When foliage is the marketable plant part, substantial losses in quality and marketability of the crop result from the injury produced by the joint action of pollutants. The amount of foliar injury affects the amount of photosynthate produced by the plant. Thus, in many instances, foliar injury provides some indication of the potential for loss in weight, size and number (yield) of the marketable plant part.

MENSER and HEGGESTAD (1966) studied the exposure of a sensitive 'Bel W_3' cigar wrapper tobacco to mixtures of O_3 (60 ug/m^3) and SO_2 (appr. 700 ug/m^3) for 2 and 4 hour periods and detected 23 percent and 48 percent foliar injury, respectively; but no visible injury was produced by the same concentrations of the pollutants individually. Later, the additive and frequently synergistic foliar injury response of tobacco was reported as occuring in numerous tobacco cultivars and types (MENSER and HODGES (1970, GROSSE ET AL. (1971), and HODGES ET AL. (1971)).

They confirmed that when the gases were combined and the plants were exposed for the same length of time, foliar injury was produced.

Since the first reports, the effects of mixtures of ozone and SO_2 have been studied using a variety of plant species. Radish and alfalfa plants showed more than additive foliar injury from a 4-hr exposure to a mix of 200 ug/m^3 O_3 and 270 ug/m^3 SO_2 (TINGEY ET AL., 1973). Greater than additive foliar injury effects have also been reported for broccoli and tobacco, while additive or less than additive effects have been noted for cabbage, tomato, lima bean, bromegrass, spinach, onion, and soybean (TINGEY ET AL., 1973). Soybean has exhibited non-significant less than additive foliar injury effects (TINGEY ET AL., 1973).

Field-grown soybeans (cv. Dare) exposed to 200 ug/m^3 O_3 alone or 200 ug/m^3 O_3 and 270 ug/m^3 SO_2 for 6 hr/day for 133 days in field chambers exhibited injury and defoliation. Injury and yield due to the mixture were increased (9 percent) and decreased (19 percent), respectively, compared to the ozone-alone treatment, but the differences were not significant (HEAGLE ET AL., 1974).

Alfalfa exposed in closed field chambers to low levels of ozone and sulphur dioxide individually and in combination for varying periods of time exhibited significant reductions in yields, quality, and nitrogen fixation compared with the control plants, but there were no significant interactive effects (NEELY ET AL., 1977).

Other sutdies have included the apple (SHERTZ ET AL., 1980), grape (SHERTZ ET AL., 1980), radish cucumber, and soybean (BECKERSON AND HOFSTRA, 1979), and pea (OLSZYK and TIBBITTS, 1981). Although relatively high O_3 and SO_2 concentrations were used for only a few hours, most species displayed a synergistic injury response from the joint effects of the pollutants, supporting previous observations.

Most research examining the effects of pollutant mixtures has utilized standard means comparisons to express the responses. These tests usually do not adequately evaluate the interaction due to the failure of one pollutant to be consistant at different concentrations of the second pollutant.

REINERT and NELSON (1980) utilized sums of squares partitioning by factorial analysis to examine the effects of 1350 ug/m^3 SO_2 and 500 ug/m^3 O_3 (4-hr exposures, 4 times, 6 days apart) on Begonia. A significantly less than additive effect was found for flower weight of 1 to 5 cultivars. The same technique was utilized by REINERT and WEBER (1980) to evaluate the effects of 500 ug/m^3 O_3 and 675 ug/m^3 SO_2 (exposed 4 h/day, 3 days/wk, for 11 wks) on soybean (DARE); an additive effect of the pollutant mixture was demonstrated.

Many studies have been conducted on the effects of mixtures of sulphur dioxide and ozone on eastern white pine (COSTONIS, 1973; DOCHINGER and HECK, 1969; HOUSTON, 1974; HOUSTON and STAIRS, 1973). Genetic control of sulphur dioxide and ozone tolerance in this species has been demonstrated, for low concentrations of SO_2 (675 ug/m^3) and O_3 (100 ug/m^3) for only 6 hrs, to consistently injure the exposed sensitive clones (HOUSTON and STAIRS, 1973). HOUSTON (1974) later used mixtures of sulphur dioxide and ozone doses to simulate actual field conditions and reported that even the lowest concentrations of O_3 (100 ug/m^3) and SO_2 (135 ug/m^3) for 6 hrs in mixture caused more serious damage than that resulting from either pollutant alone at similar concentrations. A less than additive effects on foliar injury was noted when Scotch pine trees were exposed to 675 ug/m^3 SO_2 and/or 280 ppm or 580 ug/m^3 O_3, 6 hrs/day for varying time periods (NEILSON ET AL., 1977).

Exposure of aspen clones to 100 ug/m^3 O_3 and/or 540 ug/m^3 SO_2 for 3 hrs resulted in a more than additive number of plants in the mix exhibiting foliar injury (KARNOSKY, 1976).

The chronic effects of the joint action of O_3 and SO_2 on the growth of radish, alfalfa, soybean, and tobacco were summarized in 1978 (U.S. ENVIRONMENTAL PROTECTION AGENCY, 1978). These four species represent a diverse group of plant species in terms of growth habit. Primary focus in earlier studies was on weight changes during the vegetative stage of growth, with the exception of one study (HEAGLE ET AL., 1974); however, radish root (hypocotyl), tobacco leaf weight, and alfalfa foliage (top) weight are the marketable portions of the plant. With the

exception of alfalfa foliage (top) weight is the marketable portion of the plants. With the exception of alfalfa, the growth of each plant species was reduced in an additive manner by the joint action of the two pollutants. Soybean root (fresh weight) responded synergistically to the combined effect of O_3 and SO_2 in one study (TINGEY ET AL., 1973):

The above data were obtained in greenhouse studies (except for HEAGLE ET AL., 1974). These data provided preliminary evidence that the combined effect of O_3 and SO_2 at concentrations of 0.05 ppm and greater caused an additive reduction in plant yield. In additional studies more emphasis has been given to the influence of pollutant combinations on yield (weight, size and numbers) as a measure of plant response, including the yield of flower, fruit, and seed portions of the plant. SHEW ET AL. (1982) exposed tomato to 400 ug/m^3 O_3 and 540 ug/m^3 SO_2 alone and in mixture and demonstrated that the combined effect of O_3 and SO_2 was synergistic, decreasing the weight of the largest fruit in each tomato cluster, but that the synergistic effects did not influence total fruit weight per plant.

The joint action of O_3 and SO_2 on the growth and yield components of tall fescue was studied by FLAGLER and YOUNGER (1982). Fescue was exposed to O_3 concentrations of 0.0, 200, 400 and 600 ug/m^3 and 0.0 and 270 ug/m^3 SO_2 for 6 hours per day, once a week for 12 weeks. The effect of SO_2 in the presence of increading concentrations of O_3 caused additive decreases in fescue total dry weight, root dry weight and root-to-shoot ratio. For example, O_3 decreased total dry weight 49 percent at 0.3 ppm O_3; in the presence of SO_2 there was an additional 11 percent loss in total dry weight. Ozone and SO_2 acted synergistically to decrease the number of tillers in fescue but the synergism depended on the O_3 concentration. These studies were done in a charcoal-filtered-air greenhouse in CSTR exposure chambers.

Recently, studies of the combined effect of O_3 and SO_2 have been conducted in open-top field chambers (HEAGLE ET AL., 1983; and OSHIMA, 1978).

In these experiments, O_3 levels near ambient, as well as increasing O_3 concentrations above ambient, were used in combination with two or more concentrations of SO_2. HEAGLE ET AL. (1983) exposed soybean to various concentrations of O_3 for 7 hrs daily and 4 concentrations of SO_2 for 4 hours per day. Both gases were added for 111 days. The high concentration of SO_2 decreased the amount of visible injury from increasing concentrations of O_3. The joint action of O_3 and SO_2 on soybean seed weight per meter of row at lower concentrations appeared to be additive, but as the concentrations of both pollutants increased there was an antagonistic $O_3 + SO_2$ interaction. The nature of the combined effect was similar to that for visible injury: as SO_2 increased to 1000 ug/m^3 the loss of seed weight from increasing O_3 concentrations was less than at lower concentrations of SO_2.

HEGGESTAD and BENNETT (1981) exposed three cultivars of bean to increasing concentrations of SO_2 for 6 hours per day in charcoal-filtered and unfiltered ambient air, using open-top field chambers. The beans were exposed daily, 5 days per week, for30 days. Sulphur dioxide (800 ug/m^3) reduced snap bean yields (all cultivars) in nonfiltered air (O_3) by 44 percent compared to a 16 percent reduction in charcoal-filtered air. At 160 ug/m^3 SO_2, the yield of cv. 'Astro' was reduced more in nonfiltered than in filtered air.

In southern California, OSHIMA (1978) and FOSTER ET AL. (1983a,b) conducted studies to determine the combined action of SO_2 and photochemical oxidants. A range of photochemical oxidant concentrations was obtained by combining various proportions of charcoal-filtered ambient air which was added to the field exposure chambers. Sulphur dioxide (0.0 or 270 ug/m^3) was added to the chambers for 6-hour periods approximately 47 times over a 76-day period for beans (OSHIMA, 1978) and 4 to 5 days per week over a 10-week period for potato (FOSTER ET AL., 1983). The kidney bean yield was less in the presence of ambient oxidant plus SO_2 except at the high oxidant concentrations, when the yields were more similar. Similar studies with potatoes exposed to SO_2 and partially filtered ambient air containing O_3 resulted in no evidence of joint action on tuber yield (FOSTER ET AL., 1983).

II.2.2. Combined effects of O_3 and NO_2

Although the effects of NO_2 and O_3, alone and in mixture, have not generally been studied, recent reports comparing two- and three-pollutant mixture treatments include NO_2 plus O_3 combinations. KRESS and SKELLY (1982) have studied the responses of seven tree species to NO_2 (190 ug/m^3) and O_3 (200 ug/m^3) alone and in mixture for 6 hours per day, for 28 consecutive days (Table II.2.2.-1). Virginia and loblolly pine growth, as measured by plant height, was significantly suppressed by the O_3 + NO_2 treatment, but not by the individual pollutants. Nitrogen dioxide alone significantly suppressed root and total dry weight of sweetgum; however, the joint action of O_3 + NO_2 was antagonistic on sweetgum root and total dry weight and white ash root dry weight.

II.2.3. Combined effects of SO_2 and NO_2

TINGEY ET AL. (1971) found that neither 3760 ug/m^3 NO_2 nor 1310 ug/m^3 SO_2 alone caused foliar injury. However, a mixture of 188 ug/m^3 NO_2 and 270 ug/m^3 SO_2 administered for 4 hours caused foliar injury to pinto bean, radish, soybean, tomato, oats and tobacco. Exposure to 282 ug/m^3 NO_2 in combination with 262 ug/m^3 SO_2 for 4 hours caused greater foliar injury. Traces of foliar injury were observed at 94 ug/m^3 NO_2 and 131 ug/m^3 SO_2.

MATSUSHIMA (1971) observed more leaf injury on several plant species from a mixture of NO_2 and SO_2 than that caused by each pollutant alone. He also tested different sequences of exposure. When NO_2 exposure preceded SO_2, the degree of injury was similar to that resulting from individual exposures to either gas. But when SO_2 exposure was followed by NO_2 the degree of leaf injury increased as would be typical of simultaneous exposures to both pollutants. FUJIWARA ET AL. (1973) found greater-than-additive effects when peas were exposed to 188 ug/m^3 NO_2 in combination with 262 ug/m^3 SO_2. When 376 ug/m^3 NO_2 and 524 ug/m^3 SO_2 were used, the effect was only additive.

When a large number of desert species were exposed to either SO_2 or combinations of SO_2 and NO_2 (ratio approximately 4:1) injury from SO_2 and mixtures of SO_2 + NO_2 was similar (HILL ET AL., 1974). NO_2 decreased the foliar injury threshold of SO_2 on tomatoes, geranium and petunia (DE CORMIS and LUTTRINGER, 1976). Exposure to 790 ug/m^3 SO_2 caused no foliar injury but the same concentration of SO_2 in conjunction with 940 ug/m^3 NO_2 caused foliar injury. Injury from the gas mixture increased with increasing humidity.

Observations in the vicinity of an arsenal emitting low concentrations of both NO_2 and SO_2 found foliar injury on several conifer species which was attributed to the interaction of the 2 gases (SKELLY ET AL., 1972). The maximum observed 1-hour concentration of NO_2 was 1100 ug/m^3 and 2-hour maximum for SO_2 was 1810 ug/m^3. Effects on growth rate of the two tree species were correlated with production activities at the arsenal over a 30-year period suggesting that mixtures of NO_2 and SO_2 may reduce plant growth (STONE and SKELLY, 1974).

BENNET ET AL. (1975) studied the effects of NO_2 and SO_2 mixtures on radish, swiss chard, oats and peas. Treatments consisted of 1- and 3-hour fumigations with the pollutants separately and with SO_2 and NO_2 (1:1) mixtures in concentrations ranging from 330 - 2620 ug/m^3 SO_2 and 230 - 1880 ug/m^3 NO_2. No visible injury occurred on experimental plants treated with NO_2 alone or from exposures to SO_2 concentrations of less than or equal to 1310 ug/m^3. The minimum exposure doses which caused visible injury to radish leaves were 1-hour exposures to a mixture of 940 ug/m^3 NO_2 and 1310 ug/m^3 SO_2 or to 1950 ug/m^3 SO_2 alone. The data indicated that SO_2 and NO_2 in combination may enhance the phytotoxicity of these pollutants, but relatively high doses were required to cause injury.

REINERT ET AL. (1975) summarized the effects of gas mixtures on foliar injuries to a number of crops (Table II.2.3.-1). The data indicate that ambient concentrations of NO_2 and SO_2 may interact to injure vegetation.

A study was conducted to determine the effects of low concentrations of NO_2 and SO_2 singly and in combination on the growth of four grass species (ASHENDEN, 1979; ASHENDEN and MANSFIELD, 1978; ASHENDEN and WILLIAMS, 1980). Plants were grown and exposed in four small greenhouses which received either charcoal filtered air, 210 ug/m^3 NO_2, 290 ug/m^3 SO_2 or a mixture of both gases at these same concentrations. The plants were exposed for 103.5 hours per week, which resulted in weekly mean concentrations of 130 ug/m^3 NO_2 and 180 ug/m^3 SO_2. The plants were harvested monthly and various growth parameters were measured. Nitrogen dioxide significantly reduced growth parameters of orchard grass and Kentucky blue grass, but had no effect or slightly stimulatory effect on the growth of Italian ryegrass and timothy. However, growth parameters of all the species were reduced by SO_2. The combination of NO_2 and SO_2 significantly reduced the growth parameters of all species tested and many of the effects were determined to be synergistic. These data were collected during the winter when the plants were in a period of slow growth which may have increased the pollutant's toxicity. However, the data clearly show that intermittent exposures to ambient concentrations of NO_2 and SO_2 singly and in combination can significantly depress yield parameters of important forage grasses.

Alfalfa exhibited a greater-than-additive response, i.e., a greater inhibition of apparent photosynthesis (CO_2 uptake) when NO_2 and SO_2 were applied together for 2 hours at 470 ug/m^3 and 655 ug/m^3 SO_2 (WHITE ET AL., 1974). A mixture of 282 ug/m^3 NO_2 and 393 ug/m^3 SO_2 for 2 hours decreased apparent photosynthesis 7 percent more than when the total of the two gases was applied independently. At higher concentrations the effects were not greater-than-additive.

Exposures of alfalfa to 620 ug/m^3 NO_2 and 2620 ug/m^3 SO_2 at an ambient CO_2 concentration for 1 to 3 hours reduced the photosynthetic rate approximately 50 percent (HOU ET AL., 1977). In studies with pea BULL and MANSFIELD (1974) reported that over the concentration range of 0 - 470 ug/m^3 NO_2 and 0 - 655 ug/m^3 SO_2, photosynthesis was inhibited. The duration of exposure was not given.

The effect of the two gases was additive in inhibiting of photosynthesis. In bean when 188 ug/m^3 NO_2 and 262 ug/m^3 SO_2 were applied individually the pollutants stimulated short-term increases in transpiration, but the combination of NO_2 and SO_2 decreased the transpiration rate (ASHENDEN, 1979).

HORSMAN and WELLBURN (1975) studied the effects of NO_2 and SO_2 mixtures on several enzyme systems in peas. Peroxidase activity was enhanced somewhat by SO_2 alone but not by NO_2. However, 188 ug/m^3 NO_2 plus 524 ug/m^3 SO_2 for 6 days increased the activity by 24 percent. A 100 percent increase occurred when 188 ug/m^3 NO_2 plus 5240 ug/m^3 SO_2 was used for 6 days. The effect was much greater-than-additive. The increase in peroxidase activity was considered a typical stress response of the plant. Similar studies have shown that glutamate dehydrogenase activity is stimulated as greater-than-additive response by mixtures of NO_2 and SO_2 (WELLBURN ET AL., 1976).

Very recently an attempt was made to relate the dose of SO_2 + NO_2 mixtures to effects on the growth of Poa pratensis (WHITMORE, 1985). Experiments were conducted under closely controlled environmental conditions and plants were exposed, from emergence, to mixtures of SO_2 + NO_2, in 1:1 ratio, for periods of 20, 34 and 38 days. Controls were grown for comparison in air containing approximately 19 ug/m^3 SO_2 and 14 ug/m^3 NO_2. The three exposure concentrations were 110 ug/m3 SO_2 and 77 ug/m^3 NO_2 and two higher concentrations (135 - 192 ug/m^3 NO_2, 190 - 270 ug/m3 SO_2) of each gas. In general the response (plant dry weight) increased with respect to time and concentration. It is noteworthy that exposure to only 110 ug/m^3 SO_2 + 77 ug/m^3 NO_2 for 34 - 38 days severely inhibited dry weight gain; these plants weighed only 60 - 62 % of controls.

In more recent research at Lancaster (UK), dose response studies have been performed for other species exposed to NO_2 + SO_2 mixtures. PANDE and MANSFIELD (1985) exposed 2-day-old plants of spring barley to a range of concentrations of 77 ug/m^3 - 270 ug/m^3 NO_2 and 110 ug/m^3 - 380 ug/m^3 SO_2. The data showed that, at least under these conditions, barley is less affected by NO_2 + SO_2 than was Poa pratensis.

Research on woody plants has suggested that the dose-responses to treatments with SO_2 and NO_2 may be very similar to those found in herbaceous plants (FREER-SMITH, 1985).

FREER-SMITH (1985) concluded, for Betula pendula, that the effects of a dose near a threshold of 90 - 135 ug/m^3 NO_2 and 135 - 190 ug/m^3 SO_2 could be stimulatory or inhibitory depending on the growth rate of the plants. This threshold for injury could fall from 77 ug/m^3 NO_2 and 110 ug/m^3 SO_2 when the plants were growing slowly in low light intensities.

Although information on long-term effects of this gas mixture has begun to appear in the last 4 or 5 years, it is nevertheless impossible to give precise information on threshold levels of exposure with respect to concentration or duration of treatment. POSTHUMUS (1982) did, however, achieve some success with concentration/time models and plotted "non-effect" lines both for NO_2 on its own and NO_2 + SO_2, as shown in Figure II.2.3.-1.

These models involve some speculation because of the lack of sufficient data, and this is specially true for NO_2 in combination with SO_2. Nevertheless the exercise is valuable because (a) it helps us to visualise a complex situation (b) it provides a preliminary assessment of "non-effect" levels and (c) it provides a model which can be updated as the results become available from future research.
On the basis of POSTHUMUS's model, and using the more recent experimental evidence for both herbaceous and woody species, we can deduce that when NO_2 is present with approximately the same concentration of SO_2, inhibition of growth will occur in sensitive species at concentrations upwards of 20 ppb. (38 ug/m^3 NO_2, 54 ug/m^3 SO_2). At lower concentrations, or for these concentrations at shorter exposures, growth may be stimulated by the pollution.

The non-adverse effect levels derived from POSTHUMUS's isometric projection for NO_2/SO_2 exposures are shown in the Table II.2.3.-2.

II.2.4. Combined effects of SO_2 and SPM

Particulate emissions have their greatest impact on terrestrial ecosystems near large emission sources. Particulate matter in itself constitutes a problem only in those few areas where deposition rates are very high. Ecological modification may occur if the particles contain toxic elements, even though deposition rates are moderate. Solubility of particle constitutents is critical, since water-insoluble elements are not mobile within the ecosystem. Most of the materials deposited by wet and dry deposition on foliar surfaces in vegetated areas is transferred to the soil where accumulation in the litter layer occurs.

Very few studies have reported that combinations of particulate matter and SO_2 (or particulate matter and other pollutants) increase foliar uptake of SO_2, increase foliar injury of vegetation by heavy metals and reduce growth and yield. Because of the complex nature of particulate pollutants, conventional methods for assessing pollutant injury to vegetation, such as dose-response relationships, are poorly developed. Studies have generally reported vegetational responses relative to a given source and the physical size or chemical composition of the particles. For the most part, studies have not focused on effects associated with specific ambient concentrations. Coarse particles such as dust directly deposited on the leaf surfaces result in reduced gas exchange, increased leaf surface temperature, reduced photosynthesis, chlorosis, reduced growth, and leaf necrosis. Heavy metals deposited either on leaf surfaces or on the soil and subsequently taken up by the plant can result in the accumulation of toxic concentration of the metals within the tissue.

II.2.5. Combined effects of O_3, NO_2 and SO_2

In the last few years experiments have been designed to study the effect of increasing concentrations of NO_2, SO_2 and O_3 in mixture (Table II.2.5.-1). REINERT and GRAY (1981) exposed radish plants one time for 3 or 6 hours to 0.2 or 0.4 ppm of NO_2, SO_2 or O_3 or combinations. They found no interaction for either two- or three-gas mixtures, even though the decrease in hypocotyl weight by O_3 was further reduced by NO_2 alone, SO_2 alone, or NO_2 plus SO_2, which suggests an additive response. REINERT and SANDERS (1982) and SANDERS and REINERT (1982) reported similar results in radish following repeated exposures at different ages.

Marigold was exposed at different ages for 3 hours to 0.3 ppm of each pollutant, three times per week for 1 week (SANDERS and REINERT, 1982). Ozone alone decreased flower dry weight but the interaction of NO_2 or O_3 with SO_2 was apparently antagonistic. Similar results were reported for marigold exposed repeatedly 3 days a week for 3 weeks. REINERT and HECK (1982) exposed snap beans 27 times intermittently for 3 hours each time over 6.5 weeks to increasing concentrations of SO_2 and NO_2 in the presence of 100 ug/m^3 O_3. Ozone alone decreased bean pod weight 10 percent, while NO_2 at 190 ug/m^3, SO_2 at 400 ug/m^3, and at O_3 100 ug/m^3 decreased pod weight by 31 percent. REINERT and HECK (1982) exposed 16-day-old radish plants one time for 3 hours to three concentrations or of NO_2, SO_2 and O_3 at all 27 treatment combinations. In both experiments, the reduction in size of radish hypocotyls was predominantly additive and linear within the range of concentrations used. The above studies were conducted primarily under greenhouse conditions but some of the species studied, such as marigold, tomato, and azalea, are grown commercially in greenhouses. The concentrations of SO_2 and NO_2 were below the concentration of each pollutant individual that causes visible injury for a single exposure (TINGEY ET AL., 1971).

Several turf grass species and cultivars were exposed to O_3, SO_2, and NO_2 individually, and to the combination of the three pollutants to determine the effects on leaf area (ELKIEY and ORMROD, 1980). The three-pollutant combination reduced the leaf area of only 4 of the 12 Kentucky bluegrass cultivars. The

three-pollutant combination had no significant effect on red top, creeping bentgrass, and colonial bentgrass, but it did significantly reduce the leaf area of perennial ryegrass and one of the two red fescue cultivars.

A study by MOOI (1984) at Wageningen has shown that combinations of SO_2, NO_2 and O_3 may be very damaging for both herbaceous and woody plants. Effects on Plantago major are shown in Figure II.2.4.-1.

It will be seen that the combination $NO_2 + O_3 + SO_2$ was always damaging, but there was a variable reaction to $NO_2 + SO_2$.

II.3. Combined effects on materials

The atmospheric attack of materials is partly due to air pollution. The processes of attack proceed slowly and a complex interaction occurs between material, environmental factors and air pollution. Material damage is often explained as a synergistic action of pollutants and natural environmental influences, e. g. in connection with NO_2 the most important synergetic factors are humidity and UV-radiation of sunlight. However, effects of combined action of air pollutants have been established in only a very few cases. (Table II.3-1 summarises the types of damage that can be caused by air pollution and other environmental factors).

In conclusion, it can be said that

- very little is known about combined effects of air pollutants to materials
- sulfur oxides figure prominently as a pollutant type which can cause damage to many materials.
- air pollution is only one of the environmental factors that can cause damage to materials exposed to outdoor atmospheres.

Correlation of published free-field-weathering and laboratory experiments is not easy to show due to unknown pollutant concentration or unrealistic pollutant concentration within the labs, respectively.

Even in the cases where two or more air pollutants attack materials the processes of attack are different and independent from each other so that no combined effect occurs. An exception is the attack of metals and some building stone due to acid substances, because there is a clear additive effect. However, the most abundant air pollutant which can produce acids is SO_2 and it can be assumed that in comparison with SO_2 the impact of other air pollutants is negligable, e. g. considering the

direct chemical reaction of SO_2 and NO_2 on materials such as zinc, copper and limestone ($CaCO_3$), 1 ug of SO_2 can be assumed to produce the same effect as 3.7 ug of NO_2 on the basis of differing deposition rates of the two gases and the stoichiometry of the reactions. Given the same concentration of these two air pollutants the respective proportions of damage caused by the dry deposition of SO_2 and NO_2 are 80 % and 20 %.

Moreover, there are several fundamental problems in quantifying the extent of damage to materials from air pollutants, e. g.

- Reduction in ambient levels of pollutants such as SO_2 as the result of air pollution control efforts has significantly reduced the exposure and degree of possible damage to exposed materials. Damage identified and measured today may have been the result of much earlier exposure when concentrationswere much higher.

- Changes over recent years in the types of materials and protective coatings exposed to the atmosphere as a result of technological improvements is having a fundamental effect upon the nature and extent of air pollution-induced damage to materials and associated damage costs.

- Determination of the quantities of materials in place in relation to air pollutant exposures is a difficult task, especially when significant amounts of certain materials (e. g., fabrics, paints and paper) may be exposed to indoor environments or other locations not represented by typical ambient monitors.

Therefore the damage of materials is not considered as a specific reason for studying abatement strategies for combined effects of air pollutants.

II.4. Conclusions

From the review shown in chapters II.1. to II.2. the following can be concluded:

1. Controlled human exposure studies have not consistently demonstrated any worsening of respiratory effects for combined exposures of O_3, SO_2, NO_2 and CO. There is still some concern that combinations of pollutants may contribute to the symptom aggravation and decreased lung function as described in epidemiological studies. However, for the purposes of this study, the current knowledge does not leas to a definition of any working hypothesis with regard to combined effects of these pollutants on human health.

2. There is no doubt that the interaction of SO_2 and particulate matter is an important synergistic factor in respiratory effect studies. The epidemiological data available makes it possible to define, as done by WHO, clear guidelines for the protection of human health against combined attack of SO_2 and black smoke. The WHO recommendations are:

	Concentration in ug/m^3	
	SO_2	Black Smoke
24 h mean	100 - 150	100 - 150
Annual arithmetic mean	40 - 60	40 - 60

3. The present understanding of how pollutant combinations influence plant growth and development, and how environmental factors can modify those responses is still fragmentary. However, data have demonstrated that interactions can occur between O_3 and SO_2, SO_2 and NO_2 and SO_2, NO_2 and O_3.

4. As far as combined actions of O_3 and SO_2 are concerned, effects on foliar injury and yield have been demonstrated. The observations were significant enough to propose the following general concepts:

i. When concentrations of O_3 and SO_2 are below or at the threshold for visible injury, synergistic interaction may be a frequent occurence.

ii. As concentrations of O_3 and SO_2 increase in mixture above the injury threshold, yield loss from joint action may be additive.

iii. When both pollutants are present in high concentrations, the joint action of O_3 and SO_2 may be antogonistic, such that further weight loss is minimal.

The concentration level at which combined effects may occur is in the range of 100 ug/m^3 O_3 and 135 ug/m^3 SO_2. However, additional information concerning pollutant dose and frequency of exposure, at which these effects, take place is needed. Therefore, no specific working hypothesis for the interaction of SO_2 and O_3 can be defined for the purposes of this study.

5. Synergistic effects of NO_2 and SO_2 in damaging plants have been well established for short term exposures, and recent experimental evidence suggests that synergism of the same kind can also occur in long term treatments. It is possible to establish dose-response curves for NO_2 plus SO_2 treatments for plants fumigated in controlled environment conditions. However, it must be recognised that studies have so far been performed on very few plant species, and that there is evidence that environmental conditions can greatly alter the response to NO_2 plus SO_2. Thus a concentration which is damaging in winter may have little effect in summer, or may even be beneficial. Nevertheless for the purposes of this study the following combined concentrations are taken as non-adverse effect levels:

	Concentration in ug/m^3	
	SO_2	NO_2
24 h mean	150	300
Annual average	40	40

It has to be noted that the given 24 h mean value is pure guesswork, because according to the review a combination of 250 ug/m^3 SO_2 plus 40 ug/m^3 NO_2 for 15 hours exposure time is as damaging as the combination of 40 ug/m^3 SO_2 plus 500 ug/m^3 NO_2 for 38 hours exposure time. The value given above is therefore a concentration/time average.

6. Some recent experiments in which O_3 has been added to NO_2/SO_2 mixtures suggest that the threshold for injury may be even lower. As indicated under point 5, data suggest that this threshold may be for long term exposure as low as 30 ug/m^3 NO_2 in combination with 60 ug/m^3 SO_2 and 30 ug/m^3 O_3.

   Therefore for the purposes of this study a non-adverse effect level of

	Concentration in ug/m^3		
	NO_2	SO_2	O_3
annual average	30	60	30

   will be laid down as working hypothesis.

7. The review of combined attack on materials gave no sound basis for any working hypothesis suitable for this study.

8. An important question is whether or not the combined non-adverse effect levels are more stringent than the non-adverse effect levels for the single compounds. The following overview shows such levels for single compounds:

	Non-adverse effect levels in ug/m^3 for the protection of	
Compound	Human Health	Vegetation
SO_2	effects due to the impact	50 ... 120 as annual mean
Black Smoke	impact of single compounds less important than of combined effects	none

NO_2	190 ... 320 as 1 h maximum level	4000 ... 9000 as 1/2 h value
		250 ... 500 as 7 months average
O_3	120 as 1 h maximum level	300 ... 1000 as 1/2 h value
		50 ... 300 as 4 h average

The comparison with the combined non-adverse effect levels, mentioned above, shows that

with regard to SO_2: the single non-adverse effect levels for SO_2 alone is more or less equal to the levels for combined effects as for the annual mean levels are concerned, but less stringent for the short-term effects

with regard to NO_2: the single short term non-adverse effect level for the protection of human health is more stringent than the one for combined effects on vegetation, while in the case of the long term non-adverse effect levels the one for combined effects is much more stringent than the one for NO_2-effects alone.

with regard to O_3: the non-adverse levels for combined effects are not comparable to those for O_3 alone because effects due to the combined impact of air pollutants happen during long term periods while those for O_3 alone within short term periods.

9. Generally it can be said that the uncombined limit values, as laid down in industrialized countries and shown in Tables I.2.-1 to I.2.6, are higher than the values established in chapter II.4. However, there are a few exceptions.

For SO_2:
- the Canadian and the Russion long term values are of the same order of magnitude as the one proposed in point 2.
- the class I limit values as laid down in China are more stringent than those mentioned in chapter II.4. point 2.

- the proposed limit values under discussion in Switzerland are more stringent than those taken here as working hypothesis.

For NO_2: - The Canadian, Russian, Dutch and Japanese short term values are lower than the one proposed in point 5
- the limit values proposed by the Swiss government to the Parliament are much lower than the value of point 5.

However, none of these low values are more stringent than those proposed for combined effects of NO_2, SO_2 and O_3 in point 6.

For O_3: - the annual arithmetic mean value, as laid down in Canada, is equal to the one proposed under point 6

As can be seen from these examples, there are already a number of proposals for ambient air limit values and some implemented values which are lower than those mentioned above.

10. However, even if the single limit values are higher, they may be more stringent than the lower combined values, because the exceedance of two lower limit values at the same time may be less likely than the exceedance of a higher single value. All depends on the actually chosen value and the local situation. The final answer about the stringency of one or the other set of limit values can in most cases only be answered by a comparison of actually measured data.

11. If one would follow the most stringent approach, i.e. to lay down limit values which protect human health and the environment (except against wet deposition) against single and combined effects the following concentrations should be appropriate (1):

	Long term value		Short term value	
	human health	environment	human health	environment
SO_2	40	40	100	250
NO_2	none	30	200	300
O_3	none	30	120	100
Black Smoke	40	none	100	none

(1) Very recently, the WHO Regional Office for Europe reviewed effects of air pollutants on the environment and concluded:

Recommendations

SO_2

The maximum annual mean SO_2 concentration should be 30 ug/m^3, with 100 ug/m^3 as the 95th percentile. This guideline may, however, be insufficient in the case of extreme environmental conditions and/or the presence of other pollutants.

NO_x

In the presence of levels of SO_2 and ozone not higher than 30 ug/m^3 and 60 ug/m^3 respectively, the atmospheric concentration of NO_2 should be no higher than 30 ug/m^3 as a yearly average of 24-hour means and no higher than 95 ug/m^3 as a 4-hour average.

Ozone/photochemical oxidants

Mean ozone concentrations should stay below 200 ug/m^3 for 1 hour, 65 ug/m^3 for 24 hours and 60 ug/m^3 for the growing season.

III. Ambient air concentrations of SO_2, NO_2, O_3 and Black Smoke

This chapter gives an overview about ambient air concentrations of SO_2, Black Smoke, NO_2 and O_3, in particular those measured within the Member States of the European Communities.

The aim of this chapter is to compare the measured values with the non-adverse effect levels given in chapter II in order to identify areas where risks of exceedances exist.

A particular problem of the comparison is that these risks can only be assumed if the combined measured values exceed combined limit values.

The available literature provides only little detailed information on the simultaneous time-patterns of two or more pollutants. Therefore, the assessment has been carried out in two steps:

- in the first step a general overview of ambient air concentrations of single pollutants is given and non-adverse effect values are compared with ranges of measured concentration. It is then assumed that a risk of exceedances exists if the upper values of each of the combined ranges are above the combined non-adverse effect levels and the known emission pattern indicates that such levels may occur simultaneously.

- in the second step, where necessary, a more detailed analysis based on data sets of actual measured values has been carried out for such cases where no final answer could be found in the first step.

Although this procedure contains still a margin of error the authors believe that this approach provides a quite reliable estimate of the probability of exceedances of the combined non-adverse effect levels in the Member States of the European Community.

III.1. General considerations

Ambient concentrations of the pollutants of concern are determined by the following factors: (1) the density of emissions sources; (2) the source characteristics such as stack height, exit volocity, and source strength (3) the local meteorological conditions (4) the local topography and surrounding buildings (5) the reaction rate in the atmosphere and (6) the removal rates by precipitation, deposition at surfaces, and other reactions. These factors interact in such a way that in urban and industrialized areas with high densities of SO_2, NO_2 and Smoke emissions, the pollutant concentrations are much higher than in surrounding rural areas. It is quite common to find gradients of the concentration of these pollutants within these urban with a central core area reporting the highest concentrations. NO_2 shows generally smaller gradients than SO_2 and Black Smoke. However, the general pattern is shown diagrammatically in Figure III.1-1.

Where SO_2 and NO_2 emissions are dominated by a single or a few point sources, the pattern of the concentrations could be different from the pattern displayed in Figure III.1-1. Depending on topography, meteorology, and source characteristics, the concentration patterns may be assymmetrical and the temporal distribution may be skewed to low mean values with a few intermittent high peaks. These differences in concentration patterns may be important to the effects experienced in exposed human populations and vegetation.

The pattern in O_3-concentrations is completely different from those identified for NO_2, SO_2 and Black Smoke because, as a secondary pollutant, O_3 concentrations depend particularly on meteorological conditions, like radiation, and the availability of other reacting compounds.

For the purpose of this study the site descriptions have been restricted to two categories: "Urban" and "Rural".

These are defined as follows:

- Urban: all the urban areas situated in zones frequented by people and under the strong influence of traffic, domestic and industrial pollution.

- Rural: areas of low population and very low traffic and not directly affected by domestic and industrial pollution.

There are, of course, differences in the detailed descriptions of sites, which may - to some extend - explain the relatively wide concentration ranges reported.

Data from stationary monitoring sites are used to estimate the air pollution. The air arriving at a fixed observation point at any time has a unique history. The aspects of this history which determine the ambient concentrations and relative amounts of the pollutants are the sources encountered along the trajectory and a variety of meteorological variables. Atmospheric reactions are functions of the concentrations of pollutants emitted to the atmosphere, temperature, sunlight, and time. Other meteorological factors, such as wind speed, vertical temperature structure, and the region's topography, affect the dispersion and dilution of both the directly emitted pollutants and the products of atmospheric reactions.

Given the complex nature of the processes which give rise to potential exposures of vegetation and human beings, one practical method of estimating these exposures is by monitoring atmospheric concentrations.

Once an air monitoring station's location is chosen, additional practical considerations arise relating to the actual placement of probes for sampling ambient air. Building surfaces and other obstacles may possibly scavenge pollutants from ambient air. For this reason, probes must be located a certain minimum distance away from such obstacles. It is important, also, that the pollutants of the parcel of air sampled have had sufficient time to undergo atmospheric chemical reactions (such as conversion of NO emissions to NO_2) characteristic of the polluted atmosphere. For this reason, and to

avoid sampling air dominated by any one source, probes must also be located some minimum distance from primary sources. However, it was not possible within the framework of this study to check whether or not the available data was measured under comparable siting conditions. Therefore, care must be taken in comparing one data set with another.

III.2. Observed atmospheric concentrations of SO_2 and Black Smoke

III.2.1 Introduction

Measurements of sulfur dioxide and Black Smoke in the ambient air have been made for a long time. Despite some imperfections in measurement methods for SO_2 (measurement of acidity instead of SO_2), a selection of the existing monitoring data are presented in order to gain an overall perspective with respect of SO_2 and Black Smoke.

Ultimately, the importance of the ambient air measurements of pollutant concentrations is in estimating harmful effects. Therefore, at the end of this chapter, the values reported are compared with the non-adverse effect levels mentioned in chapter II.

III.2.2. Background concentrations of SO_2

Although there are natural sources of SO_2 such as volcanoes, these are of minor importance. Sulfur dioxide has a rather short half-life in the troposphere. Background levels are rural sites of non-industrialised countries often below the monitor's detection limit.

It is not surprising that even in Europe the mean SO_2 concentration in some rural locations are below 1 ug/m^3. SO_2 concentrations measured in relatively high altitudes by the EMEP network in the summer period give a range of actual background levels within Europe (see Table III.2.2.-1). In Winter periods these values may raise by a factor of up to 3.

III.2.3. SO_2-concentrations in urban areas

More than 2000 SO_2-monitors are being operated at sites of the European Community and a large data base is available, too large to be presented in this chapter. Therefore, the ranges and pattern of SO_2-concentration will be demonstrated by some examples only.

An overview of the SO_2-concentration measured within the European Communities is given regularly by the European Commission within the framework of the Exchange of Information. The Table III.2.3.-1 shows measured values for 1981, indicating 50 percentiles in the range of 7 to 104 ug/m^3 and 98 percentiles in the range of 51 to 941 ug/m^3. Unfortunately, no details concerning the locations of the sites can be given because the data published by the EC are coded. However, the code allows for a clear distinction between rural and urban areas, hence all the values reported in the Table could be identified as values from urban sites.

Another source of information is the WHO-network from which results are presented in Table III.2.3.-2. The measured range for the arithmetic mean is 16 to 242 ug/m^3 and 78 to 939 ug/m^3 for the 98 % that is somewhat higher than the ranges given in previous Table, but still in very comparable ranges in magnitude.

More recent data are published by national authorities. Table III.2.3.-3 displays SO_2-values monitored in some German cities in the period from 1982 to 1984. The range measured for the annual 50 percentile values is 23 to 67 ug/m^3 and for the 98 percentile values 72 to 268 ug/m^3. The winter averages are somewhat higher and range between 23 and 99 ug/m^3.

Regional differences in SO_2 concentrations, as shown in the Tables, are not striking. In part, this is the result of the locational dependency of the monitor. It is therefore difficult to speculate on regional differences in SO_2 concentrations. Concentrations are influenced primarily by local sources and the locations of monitors

have clearly not been randomly chosen in each region. Several regions and cities are still reporting high concentrations; however, one should not infer that the reported concentration prevails throughout a city. High readings may exist at one or more monitoring sites, but there are substantial gradients across a city. Therefore, one or two urban sites do not typify an entire city, however, they give indication of the pollution situation.

III.2.4. SO_2-concentrations in rural areas

The number of SO_2-monitors being operated within the EC at rural sites is substantially lower than the one for urban areas. Therefore, the available data base is much smaller than for urban areas. Table III.2.4.-1 presents SO_2-data as measured in 1981 at rural sites in three countries of the EC.
The measured ranges are 3 to 40 ug/m^3 for the 50 percentiles and 22 to 135 for the 98 percentiles. In comparison with the values measured at urban sites they are lower by a factor of 2 to 5.

III.2.5. Temporal Patterns in SO_2-concentrations

III.2.5.1. Diurnal Patterns

In some locations SO_2-concentrations have distinct temporal patterns. These patterns depend on the variability of meteorological factors and on the variability of source emissions.

Diurnal variations in SO_2-concentrations reflect the changing disperson characteristics of the lower atmosphere and variations in mixing height. If emissions are predominantly from low-level sources such as residential and institutional space heating, the highest hourly concentrations will frequently occur at night and in the early-morning hours. At these times, low mixing height and decreased wind speeds lead to higher concentrations. During the day more vertical mixing usually occurs and wind speeds increase; this results in the dilution of low-level emissions.

In locations where SO_2 emissions from taller stacks are the major SO_2 source, a different diurnal pattern can occur. In these situations, typical of power plants and smelters, the highest concentrations usually occur in the morning hours just after sunrise. Levels can be almost zero at night if the source is emitting into a stratified region above a lower level inversion. Upon inversion breakup, when heating at the surface causes vertical mixing, an elevated plume can be mixed to the ground. Fumigation conditions lasting from several minutes to several hours can occur. In essence, even with tall stacks, inversion breakup that catches the plume and brings it to the surface can occur. So the peak-to-mean ratio is almost independent of stack height. The frequency on the other hand would most likely be less with taller stacks.

In concluding this section it can be said that the variations in hourly concentrations are influenced by source configuration and meteorological disperson. Therefore, it is difficult to generalize about the diurnal pattern of the hourly concentrations. It has been shown that, although there may be some similarities, the daily patterns in SO_2 concentrations are different for different locations and can change in time for any given location.

III.2.5.2. Seasonal Patterns

Concentrations of SO_2 display seasonal variability. The variability is most pronounced in areas in which there is strong seasonal variation in the emission-source or in meteorological conditions. Obviously, in urban areas where space heating is the major source of SO_2, the levels will be much higher during the heating season. Figure III.2.5.1.-1 illustrates just such situations for the London area. The highest monthly mean concentrations occur in the winter months.

III.2.6. Trends at urban and rural sites

III.2.6.1. Yearly Trends at urban sites

The SO_2 levels in most urban areas in Europe have improved steadily since the 1970's.
The trend data of Brussels, shown in Figure III.2.6.1.-1, serves as an example of a substantial decrease in SO_2-levels.

However, there are also urban areas in which little change has occured.

Table III.2.6.1.-1 shows trend data from several urban sites as compiled by the European Commission. Out of these 16 sites 13 show a clear decrease in SO_2 concentration while only 1 shows a clear increase. However, the steady improvement of SO_2 ambient air quality has been slowed down somewhat in recent years.

III.2.6.2. Yearly trends at rural sites

The trends for rural sites are far less clear than for urban ones.
A detailed analysis of data measured within the German network was carried out by the Umweltbundesamt (MONATSBERICHTE AUS DEM MESSNETZ 9/83) indicating that the SO_2-concentrations are decreasing at some rural sites and increasing at some others (Figure III.2.6.2.-1). A correlation analysis (see Table III.2.6.2.-1) confirms that result.

III.2.7. Black Smoke Background Concentrations

Black Smoke emissions are associated with combustion processes which take place in urban and industrial areas and create air pollution problems only in such areas.

Although no real background measurement stations exist in Europe, one can estimate that background Black Smoke values are well below the detection limit of the instruments and most likely below 1 ug/m^3.

III.2.8. Black Smoke concentrations in urban areas

More than 1200 Black Smoke instruments are being operated within the Member States of the European Communities and, consequently, a large data base is available.

An overview about concentration ranges within the EC can be obtained from the publications of national authorities and international bodies like the EC and the WHO. Tables III.2.8.-1 and III.2.8.-2 present data published by these two organisations. The detected concentration ranges are between 4 and 130 ug/m^3 for the annual averages and between 28 and 312 ug/m^3 for the 98 percentile. More recent data are published by national authorities and Table III.2.8.-3 serves as an example.

III.2.9. Black Smoke concentrations in rural areas

Black Smoke concentrations in rural areas are of little importance for the purposes of this study. However, in order to give a complete picture of the ambient air situation some values, measured in the United Kingdom, are displayed in Table III.2.9.-1. The range for the 50 percentile is 1 to 9 ug/m^3 and for the 98 percentile 3 to 64 ug/m^3, that is a factor of 4 to 10 lower than in urban areas.

III.2.10. Trends and patterns of Black Smoke

As in the case of SO_2 there is a clear trend towards lower concentrations in urban areas. This is demonstrated, as an example, in Figure III.2.10.-1 for three sites at Brussels.

Table III.2.10.-1 shows trend data from a number of urban sites located within the European Communities. Out of these 12 data sets 9 show a clear decrease of Black Smoke concentrations and only one increasing concentrations.

The pattern of Black Smoke concentrations in urban areas is quite similar to those of SO_2 so that no separate considerations are necessary.

III.2.11. Risks of exceedance of the non-adverse effects levels

A risk of exceedances for combined SO_2- and Black Smoke values exists only in urban and industrial areas because, in particular, the Black Smoke values are very low in rural areas. Therefore, the following considerations focus on data measured in urban areas of concern. In some cases high SO_2-values are accompanied by high Black Smoke values because both are emitted from the same source category (households and small combustion-installations). However, in other cases the occurence of high SO_2 readings may not be accompanied by high Black Smoke values, e.g. when traffic only causes most of the Black Smoke. However, for the purposes of this study the worst case should be considered first, that is the combined occurence of the highest readings mentioned in the Table III.2.11-1. Under such conditions one can argue that the non-adverse effect values mentioned in chapter II.4. are probably exceeded in some areas of the F.R. Germany, Ireland, Italy and the United Kingdom while there is only a small likelyhood of exceedances in France and Greece and probably no exceedance at all in the other Member States. This is only a broad estimate. However, it is sufficient to demonstrate that a potential health risk caused by the combined occurence of SO_2 and Black Smoke exists in some urban areas of the European Communities.

III.3. Observed atmospheric concentrations of NO_2

III.3.1. Introduction

In this section, examples of ambient concentrations of NO_2 are presented. The data presented are not intended to be a compendium of ambient monitoring activities. They have been summarized to give a representative picture of NO_2 concentrations in the European Communities and, in particular, to provide a rational basis for deciding whether or not the non-adverse effect levels mentioned in chapter II are a cause for concern.

The question of whether or not any "typical" seasonal and diurnal pattern of NO_2 concentrations exits is discussed.

Moreover, some trend data are presented. It is important to note that the data cited have not been derived from monitoring activities subject to a formal quality assurance program. Therefore, little can be said about the comparability of the data sets.

III.3.2. Background concentrations of NO_2

Data on background concentrations of nitrogen oxides are extremely limited. ROBINSON and ROBBINS (1972) summarized measurements of NO_2 concentrations from various locations, such as Panama, the mid-Pacific, Florida, Hawaii, Ireland, North Carolina, Pike's Peak, and Antarctica. From these data, they estimated that the mean background NO_2 concentration for land areas between 65°N and 65°S is 7.5 ug/m^3.

More recent measurements using modern methods have yielded results pointing to lower values. NOXON (1975, 1978) reports lower tropospheric NO_2 concentrations at a remote site in Colorado mountains of up to 0.20 ug/m^3. The author concludes that in the truly unpolluted troposphere the column abundance of NO_2 is less than 5×10^{14} molecules/cm^2. Assuming an effective length for the NO_2 column of about 2 km (CHAMEIDES, 1975; CRUTZEN ET AL., 1978), this implies a ground level NO_2 background concentration of less than 0.3 ug/m^3. If the length of the column is 0.5 km (RITTER ET AL., 1979) background NO_2 concentrations of 0.94 ug/m^3 would be implied. In addition, NOXON reports

that the ground level NO_2 concentration produced at distances greater than 50 km from urban centers seldom exceeds that for truly unpolluted air by more than a factor of 2. Summertime measurement carried out in the framework of the EMEP-network (see Table III.2.2.-1) showed values between 0.3 and 2 ug/m^3 NO_2.

III.3.3. NO_2-concentration in urban areas

In this section, representative examples of observed concentrations of NO_2 in recent years are presented. Unfortunately, the data base of NO_2-measurements is quite small in comparison to the one for SO_2 and Black Smoke.

Table III.3.3.-1 presents the results of measurements carried out in several urban areas of the EC in 1979 - 1981. The range of values is 27 to 76 ug/m^3 for the 50 percentile, when calculated on the basis of 1 hour-average, and 30 to 72 ug/m^3, if 24 hours averages are used. The 98 percentile varies between 78 and 219 ug/m^3 (1 h-average) or 61 and 202 ug/m^3 (24-h-averages).

III.3.4. NO_2-concentrations in rural areas

Only very few NO_2-monitors are being operated at rural sites. Table III.3.4.-1 displays results measured in three Member States of the EC. The measured ranges are 8 to 35 ug/m^3 (1 h values) for the 50 percentile and 36 to 84 ug/m^3 for the 98 percentile. That means that there are differences by a factor of 2 to 4 between the concentrations measured at urban and rural sites.

III.3.5. Temporal patterns in NO_2-concentration

III.3.5.1. Diurnal Patterns

It is not possible to identify one single diurnal pattern for NO_2 valid for the urban site or rural site categories because too many variables influence the build-up of NO_2-concentrations.

In rural areas the diurnal pattern usually varies between systematic variation of the concentration within 24 hours at all and small systematic variations with a minimum during the day time and a maximum at night. (See Figure III.3.5.1.-1). The minimum during the day time is caused by an increasing availability of reacting partners, like Ozone,within this time period. However, as shown in the Figure, the variations, in absolute terms, are quite small.

Several patterns have been observed in urban areas.

Two major factors that affect NO_2 concentrations, mobile source emissions and photochemical oxidation, have fairly consistent diurnal patterns in most urban areas. These usually contribute to the observed diurnal variation in NO_x concentrations (see Figure III.3.5.1.-2). Such a variation is typified by a rapid increase in NO_2 in the morning as the result of NO emissions and photochemical conversion to NO_2. This is followed by a decrease of NO_2 in the midmorning hours due to advection and increasing vertical dispersion and also loss of NO_2 in various atmospheric transformation reactions. Peaks in the NO_2 concentration are often observed to correspond to emissions occuring during the late afternoon rush hour. In some areas, small lunchtime maxima occur. At many sites evening peaks occur. Ground-level NO concentrations usually build up slowly during the night.

Figure III.3.5.1.-2 may illustrate such a pattern.

In the early morning hours before sunrise a significant and constant NO_2 concentration may exist, presumably carried over from the previous day. During these hours, the O_3 concentration is quite low. Nitric oxide concentrations are high, most probably the result of continuous emissions during the night and early morning hours. There is no discernible NO_2 formation. After sunrise, the NO_2 concentration increases sharply as a result of photochemical reactions. Photochemical generation of NO_2 is followed by a concomitant rapid increase in O_3 concentrations, which depresses the NO concentrations until the later afternoon hours when decreasing

radiant energy and increasing NO emissions overwhelm the ozone gernating mechanism. From about 4:00 to 6:00 p.m. there is still sufficient ozone present to oxidize NO to NO_2 rapidly in a simple titration reaction apparently not involving hydrocarbons. Significant quantities of NO are oxidized leading to high nighttime NO_2 concentrations. It may be postulated that this mechanism is operative also in other localities exhibiting elevated NO_2 levels after photochemical activity, including NO_2 photodissociation, decreases in the evening.

In the case of the absence of Ozone the pattern is somewhat different and dominated by the sources which lead to build-up of NO_2 over the day, interrupted only by periods of reduced emissions, e.g. lunch hours. The Figure III.3.5.1.-3 may illustrate such a pattern.

III.3.5.2. Seasonal Patterns

The month-to-month variations in average NO_2 concentrations may be the combined result of high photochemical activity in the summer months, time-varying emissions of NO_x (with high emissions of NO_x during the winter in some areas), time-varying emissions of hydro-carbons, and area-specific meteorological conditions throughout the year. The normal pattern for the northern parts of Europe is a marked peak in NO_2 concentrations during the winter months. (See Figure III.3.5.2.-1). In contrast to the above patterns, certain sites in southern parts of Europe (Greece, South Italy) may not exhibit a marked seasonality. The seasonal patterns for rural sites follow in principal those for urban sites, however, peaks are not as high as in urban areas and in some cases they nearly disappear.

The Figure III.3.5.2.-2 and III.3.5.2.-3 illustrate this fact by showing on one hand a clear pattern for four rural sites located in the F. R. Germany and on the other hand only indications of a pattern for another rural site (Schauinsland).

Therefore, no single seasonal pattern exists for all Member States of the EC.

III.3.6. Trends at urban and rural sites

III.3.6.1. Trends in urban areas

There are only very few data sets covering long periods of time on NO_2 concentrations. Caution is necessary in using these data because collection and reporting procedures were not subject to detailed quality assurance checks and more than one operation and maintenance procedure may have been used over the years at a given site. Nevertheless, the data collected provide a useful historical perspective on trends on NO_2 concentrations. The Figures III.3.6.1.-1 and III.3.6.1.-2 present trend data for Frankfurt (F. R. Germany) and some Dutch cities, calculated as annual averages. Trends in concentrations within the periods covered were generally upward for all these sites monitored. There is little doubt that similar trends are valid for the most of the urban sites within the EC because emissions of NO_x increased in all countries within the last few decades.

III.3.6.2. Trends in rural areas

Sophisticated statistics have to be applied in order to assess trends in rural sites because the concentrations are low and influenced by a number of parameters. The Umweltbundesamt (F. R. Germany) has carried out a trend analysis on the basis of the results obtained in the UBA network. (MONTATSBERICHTE AUS DEM MESSNETZ, UMWELTBUNDESAMT 8/84 and 9/83). Figure III.3.6.2.-1 shows the curves of different percentiles as calculated from the results measured at five different sites. In the statistical analysis it could be demonstrated that at the sites, Deuselbach, Waldhof and Schauinsland, a significant increase in NO_2-concentrations occured within the period covered. The results calculated for Brotjacklriegel and Westerwald are not significant. Table III.3.6.2.-1 displays the results of the trend analysis in more detail. These results may be also valid for other rural areas in middle and north Europe, however, the picture in the southern part of Europe may be different due to the differences in the NO_2-emission increase, the meteorological conditions and the chemical processes in the atmosphere. Unfortunately, no trend data are available for that part of Europe.

III.3.7. Risks of exceedances of the non-adverse effect levels

Table III.3.7.-1. displays the ranges of NO_2- and SO_2-concentrations measured at rural and urban sites.

A comparison of the measured values with the non-effect levels for combined effects of SO_2 and NO_2, as given in chapter II indicates that there is a risk of exceedances of the 24-hour mean values and of the annual average values in urban areas, but not in rural areas.

However, synergistic effects due to the combined action of SO_2 and NO_2 have been identified mainly for plants and the values given in chapter II.4. are such that damage may be caused to sensitive plants only.
Therefore, the question of possible exceedances in rural areas is of such great importance that in a second phase of data evaluation, analyses of times series of SO_2- and NO_2-concentrations have been carried out at a number of sites in the Netherlands and the F. R. Germany. In these checks annual data sets for different sites have been analysed in order to determine the simultaneous occurence of NO_2 and SO_2 at given concentration thresholds. Because the likelyhood of a simultaneous occurence of the 24 h non-adverse effect levels is extremely low the study of the data sets was focused on the annual thresholds only. Table III.3.7.-2 shows the results of these investigations. The percentage of days at which exceedances of the threshold 40 ug/m^3 SO_2 and 40 ug/m^3 NO_2 happened is relatively low, varying within less than 1 % to 20.5 % of the measured values. That means that this detailed analysis confirms the first estimate and that exceedances of 40 ug/m^3 NO_2 in the presence of 40 ug/m^3 SO_2 calculated as annual averages are extremely unlikely in rural areas. There remains therefore the important question, whether one should require that the non-adverse effect values should not be exceeded at urban areas . However, because in our days the plants concerned are found only in some rural areas and in extensive occurence in urban areas is hardly possible due to other constraints, e.g. the lack of space, it does not seem justifiable to require that these values have to be complied with in urban areas.

III.4. Observed atmospheric concentrations of O_3

III.4.1. Introduction

The data presented in this chapter on the concentrations of ozone in ambient air are intended to provide information on measured concentration ranges and seasonal and diurnal patterns in selected urban and rural areas. In addition, some historical data on trends in ozone concentrations are given to help put more recent data in perspective.

Ozone is routinely monitored only at relatively few stations within the EC. Consequently, no comprehensive information is available for O_3.

The concentrations of ozone (and related photochemical oxidants) observed in ambient air are the net result of a combination of any or all of a variety of atmospheric processes, including:

1. Local photochemical production from oxides of nitrogen and reactive volatile organic compounds.
2. Transport of ozone produced photochemically but not locally.
3. Intrusion into the troposphere, even to ground level, of ozone-rich air from a ubiquitous strastospheric reservoir.
4. Formation of ozone photochemically in the mid-troposphere, with subsequent intrusion into the boundary layer.
5. Chemical scavenging in the atmosphere of ozone and other oxidants; e.g., the reaction of ozone with nitric oxide (NO) or the reaction of H_2O_2 with sulfur dioxide (SO_2).
6. Physical scavenging in the atmosphere of ozone and other oxidants; e.g., the temperature-dependent decomposition of PAN, the precipitation scavenging of H_2O_2, and the photolytic dissociation of ozone.
7. Combined physical and chemical scavenging processes at the earth's surface, e.g., the deposition of ozone on reactive biological or nonbiological surfaces, such as vegetation, soils, or certain polymers.

These processes include, obviously, both manmade and natural processes and driving mechanisms. Although the occurance of high ozone concentrations is most commonly associated with recognized meteorological conditions that involve intense sunlight and elevated temperatures, the variety of processes that may be involved contribute to strong diurnal cycles, but peak concentrations have been observed to occur at almost any time of day. Ozone may be transported after its formation for distances of 1000 km or more. As a result, high concentrations of ozone occur not only near large sources of precursors but also in downwind nonurban areas. Ozone can be transported at night above the surface pollutant and nocturnal inversion layer. Thus, low morning concentrations of ozone are no measure of the potential for high concentrations later in the day because downward mixing from the transported ozone reservoir aloft will occur. During daylight hours, ozone can be transported considerable distances at or near ground level.

However, emphasis of this chapter is on documentation of concentrations rather than on explanations of possible causes of observed concentrations.

III.4.2. Background concentration of O_3

As mentioned in section III.4.1. the ozone concentrations observed at nonurban sites could be the result of either transported manmade pollutants or naturally generated ozone, or combinations of both. Thus, it is not possible to categorize nonurban sites arbitrarily as being indicative of the natural atmospheric background; in fact, many rural areas can be shown to be strongly affected by upwind urban pollutant sources.

The locations in Tables III.4.2.-1 and III.4.2.-2 are a mixture of obviously remote locations, such as Antarctica and Mauna Loa; and rural locations that may or may not be immune from urban transport problems, such as Arosa and Zugspitze.

Concentrations of ozone can be expected to vary with altitude and with latitude. These variations occur because of the interchange mechanisms involved in stratospheric-tropospheric exchange, the decay of stratospheric ozone as it traverses the troposphere and the known production in the apparently unpolluted troposphere of ozone at certain

altitude ranges above mean sea level. SEILER and FISHMAN (1981) reported on ozone measurements taken on flights in remote tropospheric air during July and August 1974. Averages of their data show that ozone concentrations increase with increasing altitude and in general substantiate the accepted belief that the earth's surface and the lower atmosphere act as ozone sinks. According to the authors, the value for the average background ozone concentration in the troposphere is about 70 ug/m^3 $\pm$ 30 percent. Tropospheric ozone shows a marked hemispheric asymmetry (see Figure III.4.2.-1).

In conclusion it can be said that there is no single background concentration for O_3 but a range which may be 30 to 80 ug/m^3 for annual averages and 80 to 120 ug/m^3 for the maximum values.

III.4.3. O_3 concentration in urban areas

Only a few O_3-monitoring stations are operated within the Member States and access to these data is sometimes difficult because they are not published regularly. Table III.4.3.-1 presents the results of measurements carried out at urban sites located in Member States of the EC. The detected ranges are 25 to 60 ug/m^3 for the annual mean and 50 to 140 ug/m^3 for the 98 % percentile.

III.4.4. O_3 concentrations in rural areas

As mentioned in the preceding section, very few ozone monitoring stations are located in the Member States of the EC. Consequently, the aerometric data base for nonurban areas is as small as that for urban areas. The nonurban data presented in this section were obtained mostly from the UBA monitoring network that is designed to provide ozone concentrations at rural sites. However, these sites do not all represent areas totally unaffected by manmade ozone or its precursors due to the transport of ozone or its precursors from upwind urban areas.

Table III.4.4.-1 displays values measured at such sites in the F.R. Germany. The observed range for the annual mean is 38 to 112 ug/m^3 and for the annual 98 percentile 98 to 204 ug/m^3.

Values reported for France are in the same range (40 to 70 ug/m^3 and 50 to 180 ug/m^3, respectively).

III.4.5. Temporal patterns in O_3-concentrations

Concentrations of O_3 follow a general seasonal and diurnal trend as it will be demonstrated in the following sections. Moreover, O_3-episodes have been detected within which concentrations above 200 ug/m^3 can last for several hours or even days.

III.4.5.1. Diurnal patterns

Diurnal patterns of ozone may be expected to vary with location, depending on the balance among the factors affecting ozone formation, transport, and destruction.

However, very often the diurnal concentrations of ozone follow more or less the same characteristic pattern in urban and rural areas. This pattern is one in which the maximum concentrations occur near midday, from the late morning to the early afternoon. Figure III.4.5.1.-1 shows examples of this diurnal pattern as measured at two urban (CLL, STE) and two rural sites (CAN, SIB) in the United Kingdom.

The peaks are higher in summer than in winter periods when they may even disappear due to the dominant emissions of NO_x and/or the absence of the photochemical cycle due to reduced radiation (see curve for CLL). This sort of pattern is usually explained by the fact that ozone-forming photochemical reactions require several hours to produce a maximum concentration and also that in mid-afternoon atmospheric dilution processes and wind transport usually reach a maximum, thus producing the afternoon decline in concentrations.

At rural sites the diurnal cycle may also result from the photochemical cycle of natural HC and NO_x precursor reactions and/or from the diurnal mixing cycle in which the deeper midday mixing replenishes the ozone concentrations at the surface from the mid-tropospheric layers. This mixing compensates for the ozone scavenged of groundlayer stability at the earth's surface during the nighttime period.

Diurnal profiles of ozone concentrations can vary from day to day at a specific site and from site to site in the same area because of changes in the various factors that influence concentrations. Such site to site variations are clearly demonstrated in Figure III.4.5.1.-2 which shows diurnal variations in ozone concentrations on 2 sites in the F. R. Germany. Differences in magnitude occur that are especially noticeable between 2:00 and 5:00 p.m. Transport and availability of reacting compounds are probably involved in these variations.

The Composite diurnal data as shown in Figure III.4.5.1.-1, often differ markedly from the diurnal cycle shown by concentrations for a specific day.

Transport has been mentioned as a factor that influences the urban diurnal cycle. This transport, by horizontal wind movement, serves to reduce the concentration in the source area and to increase subsequently the concentrations in a non-source area at a later time and in a downwind direction. Thus, when the diurnal pattern for a given site shows a peak concentration rather late in the day or at night, it is logical, though not necessarily correct, to attribute this to transport influences. However, depending upon proximity to urban centers and wind speed and direction, rural areas usually experience their peak concentrations later than those of urban areas.

It also is obvious that the maximum ozone concentrations that are observed at a given site are a complex function of precursor sources and meteorological conditions. Thus, the extrapolation of observational data from one region to another or even within a given region is not a simple task and should be approached with caution.

III.4.5.2. Seasonal Patterns

Seasonal variations in ozone concentrations have been observed for many years. In the absence of dominating anthropogenic influences peak concentrations most frequently occurred in the spring months and were attributed to the influence of seasonal changes in both the stratospheric source and vertical transport mechanisms. Figures III.4.5.2.-1 and III.4.5.2.-2 show the average monthly ozone concentrations at

Hohenpeissenberg (F. R. Germany) and Mauna Loa, Hawaii, a mountain station 3000 m above sea-level.

Mauna Loa can be considered as a background station, while Hohenpeissenberg is a rural site.

A springtime maximum is discernable for both stations.

For areas influenced by manmade sources, ozone concentrations tend to follow a long-recognized pattern, with maximum average concentrations occurring mid- to late summer. This pattern is attributable to seasonal meteorological conditions that tend to favour the accumulation of higher concentrations of pollutants and the occurence of favourable photochemical reaction conditions during the late summer months. Figure III.4.5.2.-3 shows such a pattern for three urban areas located in Switzerland. Peak concentrations occur in August showing the typical photochemically related summertime ozone cycle.

III.4.5.3. Photochemical episodes

Photochemical episodes happen on local and regional scales in Europe. Within such episodes high O_3-concentrations are built up and /or transported which may last several days. Figure III.4.5.3.-1 displays the occurence of a local episode which took place in the Roubaix area in September 1982. O_3-concentrations between 250 and 500 ug/m^3 were measured over a period of three days (DECHAUX ET AL., 1985). As an example of a European wide episode, O_3-data for the period May 30 to June 5, 1982 are presented in Table III.4.5.3.-1 for several sites in Europe indicating elevated concentrations within this large area.

III.4.6. Trends at urban and rural sites

The trend data is extremely small, so that no final answer to the question whether or not trends in O_3-concentrations exist can be given. However, some results will be presented in the following sections.

III.4.6.1. Trends in urban areas

Table III.4.6.1.-1 presents data from three urban sites in Germany. It is obvious that no trend in O_3-concentrations at all can be derived from these measurements.

III.4.6.2. Trends in rural areas

Data presented in section III.4.4., while covering a number of years, do not permit the evaluation of actual trends in ozone concentrations. Determination of whether ozone concentrations in ambient air are static, rising, or declining can only be determined over a period of years. The trend in concentrations of ozone at rural sites in Germany over the period 1976 through 1982 is shown III.4.6.2.-1 (FRICKE, 1983).

Although a small increase of O_3-concentration may be derivable from these data a clear statistical trend cannot be established because of the variations due the other parameters, e.g. seasonal pattern, influence these results too much.

III.4.7. Risks of exceedances of the non-adverse effect levels

The concentration ranges reported in the section III.2. to III.4. for SO_2, NO_2 and O_3 are summarized in Table III.4.7.-1. A comparison of the measured values with the non-adverse effect levels given in Chapter II shows that there is a large likelyhood of exceedances in urban areas and a small one in rural areas. The probability of exceedances in rural areas is of relatively greater importance than the one for urban areas because, as in the case of combined effects of SO_2 and NO_2, the plants concerned grow only in such areas.

Therefore, in a second step, a more detailed study on the probability of exceedances has been carried out using data from the Dutch and German network. Table III.4.7.-2 shows the results which indicate that the probability for exceedances of the non-adverse effect levels for combinations of SO_2, NO_2 and O_3 is extremely low. These data confirm that, as in the case of combined effects of SO_2 and NO_2, exceedances are likely in urban areas only.

III.5. Conclusions

The following conclusions can be drawn from the investigations on ambient air concentrations given in this chapter:

1. The combined non-adverse effect levels for SO_2 and Black Smoke of

	SO_2	Black Smoke
24 h mean	100 - 150	100 - 150 ug/m^3
Annual arithmetic mean	40 - 60	40 - 60 ug/m^3

are exceeded in several urban areas within the Member States of the European Community.

2. The combined non-adverse effect levels for NO_2 and SO_2 of

	SO_2	NO_2
24 h mean	150	300 ug/m^3
Annual average	40	40 ug/m^3

are probably exceeded in some urban areas, but - very probably - not in rural areas.

3. The combined non-adverse effect level for SO_2, NO_2 and O_3 of

	NO_2	SO_2	O_3
annual average	30	60	30 ug/m^3

is probably exceeded in several urban areas but not at all in rural areas.

The most important reason for the fact that exceedances of the non-adverse effect levels mentioned in points 2 and 3 in rural areas do not occur is that the SO_2-concentrations are fairly well below 40 ug/m^3, calculated as annual average, or 150 ug/m^3, calculated as 24 h mean in rural areas.

IV. STRATEGIES TO REDUCE POLLUTION BY SO_2, NO_2, HC AND PARTICULATES

IV.1. Introduction

In the previous chapter information was outlined concerning the combined effects of air pollutants, the limit values applied in industrialized countries and the measured ambient air concentrations of the pollutants.

One of the main conclusions that can be drawn from chapters I to III is that, at present, the available knowledge is insufficient to permit the formation of an overall strategy which takes into account the combined action of pollutants. It is even beyond the scope of this study to define an overall strategy for even one of these pollutants.

In the light of the information available, the following chapter can only highlight some aspects concerning appropriate control strategies.

IV.2. Principles of Air Quality Management Systems

IV.2.1. Introduction

This chapter highlights general aspects of Air Quality Management Systems and tries to describe them in a systematic approach.

It becomes more and more evident that it is not sufficient to control air pollution by means of single isolated actions; control actions have to be integrated into a coordinated Air Quality Management System (AQMS). Several Air Quality Management Systems are possible depending on a variety of political, scientific and economic constraints. These constraints include the goals of a European and national air quality protection policy, the position of air quality protection within the priority scale of public interest, the legal basis, the state and the

development of technology and the demographic and economic development. An AQMS, as described below, combines several elements with respect to legislation, implementation and control effectiveness.

An AQMS must address itself to the application of environmental assessment and control procedures concerning the vast discharges of pollutants into the environment. This effort has barely begun. Environmental legislation in the EC is diverse and varied and includes mostly regulations dealing with separate permits for discharges into water, air and soil.

Land use and air quality can be expressed as observable facts. Planned land use and desired air quality are long-term objectives which need to be attained. Both are indispensable parts of a control strategy.

They are all the more important because our knowledge of the temporal and spatial relationship between emissions and their effects, as outlined in the previous chapter, is incomplete. Temporal effects may be immediate or cumulative over long periods; spatial effects may vary from a local impact to effects over long distances.

A complete AQMS is expensive and requires considerable effort. An important aspect in this context is the relation between AQMS and employment. Beneficial and adverse effects on social and economic structural patterns will result (e.g. on population, migration, growth, product costs, public and private industry investment, etc.).

An AQMS can have a positive impact upon employment due to an increased demand for labour with various technological skills (e.g. for construction of air quality measurement instruments, telecommunications and data-processing systems, pollution abatement equipment, and manpower for government and industry control activities). In a relatively short period of time, the beneficial effects upon the social, economic and political aspects are expected to outweigh the initial adverse effects (e.g. monitoring costs, abatement equipment capital expenditures, reallocation of certain industries, etc.).

It should be noted that an AQMS is needed primarily in those regions where adverse effects caused by air pollution exist or are expected to occur in the future.

IV.2.2. Description of Air Quality Management Systems (AQMS)

An AQMS is the coordinated assembly of

- the assessment of present ambient air quality, emissions and related factors
- the comparison with existing standards, criteria and guidelines.
- projection of future emissions and ambient air quality
- the development, implementation, and revision of a control strategy including economic aspects and interactions with other environmental media.

It aims to attain and maintain an acceptable level of ambient air quality. Although emphasis is mostly placed upon human health effects, it includes all ecosystems to obtain an approach covering the whole of the environment.

The AQMS comprises the past history, the present situation and future developments of air quality in the region concerned. In the AQMS the assembled information is used to develop a strategy plan. The plan is implemented, results are analysed and, if necessary, the plan is revised within the framework of the AQMS.

To attain this goal, an AQMS can use:

- emission standards (including design, construction and operating standards)
- equipment standards and product standards
- ambient air quality standards or objectives.

Appropriate combinations of emission standards, ambient air quality standards or objectives, product and equipment standards and other principles (e.g. stand-still) produce the most complete AQMS (see Fig. IV.2.2.-1).

Its characteristic elements are:

- Emission standards defined on the basis of the best available control technology, independent of ambient air quality. The same advanced technology has to be applied in polluted as well as in unpolluted areas.

- Ambient air quality standards or objectives, defined on the basis of the relationship between a pollutant's dose, concentration, deposition rate or any other air quality related factor and the direct and/or indirect effects on receptors.

- Avoiding degradation of air quality in regions where pollution levels are below the ambient air quality standards (principle of non-degradation).

- Equipment standards and product standards as described in this guideline.

- Incorporation of air pollution aspects in all town and regional planning processes.

An AQMS using EMISSION STANDARDS limits the discharge of air pollutants from stationary and mobile sources expressed either in terms of concentrations or mass flow rates. Emission standards should be defined on the basis of the best available control technology (Fig. IV.2.2.-2).

Best available control measures should take into account advanced technology, even if this technology has been successfully applied on a pilot scale only. Application of the best available control technology can easily be enforced in the case of new plants (e.g. by means of a licensing procedure). The set of regulations to achieve suitable emission reduction is generally developed in the assessment phase and integrated into the comprehensive strategy plan. Available control technology and sound measuring techniques are absolutely essential to the successful application of emission control regulations, yet surprisingly these requirements are often ignored in the haste to reduce emissions in a community. The first generation of emission control regulations often reflects inexperience, unrealistic time schedules, inadequate data bases, and a rather idealistic belief that the objectives will be achieved simply by having them written into a regulation. Despite these difficulties there is often a remarkable similarity of emission control laws in widely diverse regions, probably due to the forced deadlines and a common need to accept advice from anyone with previous experience.

Initial efforts to reduce emissions invariably emphasize the control of existing sources and this approach is sometimes costly due to the retrofit of in-place processes and equipment already in place. As the AQMS evolves, the emphasis is shifted mostly towards preventing new emissions of pollutants through the use of emission standards. It should be recognized that the control of emissions from existing facilities usually involves compromise by both the responsible agency and the industrial facility. Thus, the final outcome is settled on a case-by-case basis and may differ significantly from the required reductions for a similar source in another area. Emission standards for new sources, on the other hand, do not have this disadvantage since they can be applied in advance and uniformly to all new sources employing a given process or producing a particular product.

Air pollution results from industrial plants as well as from the more numerous small sources, e.g. space heating devices and motor vehicles. It may therefore be necessary to set up emission standards for these source categories as well. It is extremely difficult to apply best available control technology to old heating installations or cars, consequently the use of emission standards may not be adequate.

Even if the concept of emission standards is applied, an agglomeration of industry, population and traffic, in combination with meteorological and topographical conditions, may still cause unacceptable levels of air pollution.

Other means to reduce emissions are EQUIPMENT STANDARDS and PRODUCT STANDARDS (Fig. IV.2.2.-3). Equipment standards specify design characteristics for equipment, product standards specify the composition of fuels, raw materials or products. Both types of standards are normally only applied to equipment or products used in large quantities.

Equipment standards and product standards may help to reduce emissions more generally than emission standards for single sources. For instance, a limitation of the sulfur content in coal or fuel oil will reduce SO_2-emission from all coal and oil burning facilities in a given country. Limitation of the lead content in gasoline will reduce the lead emission of all motor vehicles. Specifications for construction details of motor vehicles, e.g. the recycling of crankcase emissions, or absorbing emissions from gasoline tank pressurizing systems will reduce emissions of all cars.

To meet product standards, development of new processing techniques may sometimes be necessary; to produce fuel oil with low sulfur content, new desulfurization devices in refineries must be installed.

An appreciable reduction of air pollution by means of equipment standards or product standards may be achieved if they are applied on a nationwide or international basis.

While emission standards, equipment standards, and product standards affect sources and emissions directly, AIR QUALITY STANDARDS specify the level of ambient air quality which should be achieved (Fig. IV.2.2.-4). These standards relate directly to measurable effects on health or welfare. Frequently these ambient air quality standards or criteria lead to further emission controls of specific sources. In the USA maximum allowable fluoride concentrations in animal fodder have been successfully used as a receptor/effect standard to trigger further emission controls in aluminium processes.

For the position of ambient air quality standards within an AQMS see Fig. IV.2.2.-4. All measures affecting the total amount and the spatial and temporal distribution of emissions are related to this ambient air quality level.

Normally, this level is defined in terms of concentrations of air polluting substances which should not be exceeded during a specified time period in a specified area.

This desired level may

- be based on the relationship between the dose, concentration, deposition rate or any other air quality related factor of a pollutant and its direct and/or indirect effect on receptors,

- state that the present ambient air quality should not deteriorate (non-degradation principle),

- - in the absence of the above - state that ambient air quality should be as good as it has been x years ago,

- state that ambient air quality should be as good as it is in a certain other region.

If the desired level of ambient air quality is defined on a legal basis, it is called a standard, if not, it may be regarded as an objective.

The basic requirements of an AQMS based on ambient air quality standards or criteria are the following:

- existing ambient air quality and emissions have to be identified by means of an assessment program;

- future ambient air quality has to be projected by means of an air quality simulation model, using a projected emissions inventory together with all other relevant input parameters;

- existing and expected future ambient air quality has to be compared to standards or objectives;

- if the existing or the expected future ambient air quality exceeds the standards or objectives, a strategy plan must be set up.

The advantage of this approach is that it defines the objective of the management program on the basis of ambient air quality; all abatement measures are then designed to meet the objective.

The disadvantages are - if it is applied strictly on its own - that it may impose different emission abatement measures to the same sources in regions with different pollution levels and that it may cause a region to become saturated with pollution until the standard is reached.

IV.3. Strategy Plans

IV.3.1. Introduction

An air pollution strategy plan is a master plan that provides a solution to a municipal, state, provincial, national or international air pollution problem. A strategy plan is the final and most difficult part of an AQMS. First a strategy plan should be based on ambient air quality standards, on projected emissions and ambient air quality and on the results of the assessment studies. The ultimate decision has to include also the interactions with other environmental media (land use, water pollution, etc.) and economic aspects. The selected measures of the strategy plan must be legally enforcable, practically and technically feasible.

Because of technical and/or economic reasons it is sometimes necessary to find specific solutions for existing industrial plants. Improvement of these plants requires a longer time period and this must be taken into consideration in preparing the strategy plan.

Generally speaking the selection of the initial strategy plan should be based upon four considerations, most of which have been already described in previous chapters

- determination of air quality,
- determination of the sources of air pollutants (emissions inventory),
- modeling and projecting of air pollution emissions to forecast future air quality,
- formulation, implementation and enforcement of air pollution control.

The strategy plan must be reviewed periodically and revised as appropriate.

IV.3.2. Major Strategy Plan Options

The need for a strategy plan depends on the results of the determination of emissions, on the anticipated concentrations and the effects of the air pollutants found.

In some cases the establishment of a strategy plan can be very important if many different air pollutants occur simultaneously, causing synergistic effects or secondary air pollutants (e.g. photochemical air pollutants). At times it will not be sufficient to concentrate on criteria pollutants only, but it will be necessary to include all information available about other pollutants. The emissions inventory and the results of simulation modeling provide information on the sources of the different air pollutants. It is necessary, then, to consider major sources outside the area of concern because their emissions may be transported over long distances and impact the problem area of the AQMS.

A strategy plan must not be established only on the basis of the present situation. Future emissions must be projected taking into consideration the realistic development of industrialization, population and traffic pattern as well as future land use. Changes in land use may not only affect emission patterns but may also have an influence on local wind fields and on the intensity of transport of fresh air into a polluted area. Diffusion models can be used to predict future ambient air quality.

To be effective, a strategy plan must contain legally enforceable measures to improve ambient air quality with schedules and time tables for compliance with the plan's objectives. The measures appropriate for the area of concern will depend on the objectives of a control plan and the specific local situation. Appropriate measures are emission control, land use planning, siting of industrial plants, planning of motorways and roads, designation of green belts and buffer zones, and the development of alternative energy sources and supply systems.

If air pollution is caused mainly by stationary industrial sources, very often a significant reduction of emissions will be required to accomplish the desired results of the control plan. This can be done by technical measures such as the installation of more efficient gas cleaning devices, application of new technologies, and the usage of less polluting fuels and raw materials. In many countries a uniform procedure for granting licences under a number of different environmental protection laws is now being discussed and in some cases being implemented. In doing so, great interest has been expressed in environmental impact assessment as a potential instrument for integrated planning. In heavily polluted areas a new source should only be constructed if emissions from existing sources are reduced by an equivalent amount.

For large metropolitan areas, planning and choice of the energy system is highly important. If dust, smoke, and sulphur dioxide are the main air pollution components, a significant improvement of air quality may be achieved using natural gas instead of coal or fuel oil. The installation of a remote, centralised heating system leads to an improvement of air quality in areas dominated by low level emissions from small single space heating equipments. Other measures are necessary if air pollution is mainly caused by traffic emissions. In these cases, improvement may additionally be achieved by the restriction of traffic densities in certain areas or by providing a low emission public transport system. The construction of new motorways and roads outside the area of concern may be an effective measure for lowering traffic densities in the most polluted districts. Relocation of traffic or of industrial plants has to be planned extremely carefully. A poorly thought-out plan may not achieve a net reduction in total emissions because emissions are transferred into other areas, possibly causing new problems or aggravating existing problems.

A new procedure, adopted more and more in regional planning, is the proposal of alternatives which usually differ according to the methods of protecting the quality of the natural environment. Legislation must recognize and promote the coordination between the national, regional

and local levels of government and the procedures for citizen involvement and appeal. A small AQMS, perhaps implemented by local authorities, sooner or later becomes part of a larger regional, national or international AQMS because of the long-range transport phenomena. Several alternatives for land use must be considered in light of current and future expected air quality. All other parameters of environmental quality (water, soil, noise, etc.) have importance. But as air quality is influenced by changing meteorological conditions (and is perhaps most sensitive to long-range effects), it is also one of the most difficult parameters to control in land use planning.

Existing land use and existing air quality are facts. Long-term planned land use and long-term desired air quality are objects to be managed. Both are indispensable parts of a control strategy plan. They are all the more important because our knowledge of the temporal and spatial relationship between emissions and their effect is incomplete. Temporal effects may be immediate or cumulative over long periods; spatial effects may vary from local impacts to regional impacts.

A strategy plan should also include the installation of a monitoring system for the emissions of stationary and mobile sources, ambient air quality and relevant meteorological parameters. An information system for economic, demographic and topographic data should be established. These data are of special importance for predictions of emissions and air quality and for the revision of strategy plans.

IV.3.3. Projected Emissions and Ambient Air Quality, including Impact Statements

Detailed forecasting methods are also needed for a complete strategy. In this context the emission forecast not only provides future estimates based upon today's strategies, but also provides an ideal mechanism for comparing effects of various control strategies. Thus, emission forecasting is a vital management tool and has an integral role in the planning and evaluation activities of control agencies. A good forecasting system utilizes the most current emissions inventory data and assures that the most current data are always utilized for predictive purposes.

Dispersion models are being developed to provide mathematical relationships between air pollutant emissions and ambient air quality. These models can be used to relate the contributions from different source categories and configurations to ambient air concentrations. Consequently, they can be used to evaluate the various proposed air quality abatement strategies, viz.:

- Identify the most effective way of controlling existing emissions if it is decided that existing air quality needs to be improved,

- Quantify the effect of projected emissions, including industrial/residential growth, and the effects of emission control options.

In principle, using dispersion models, air quality can be projected for as far forward as reliable emission estimates can be made. These modeling techniques are therefore integral parts of selecting control strategies, of revision of strategy plans and of impact statements.

Most projection studies are primarily concerned with predicting the mean values of the pollutant concentrations in ambient air and the probability of occurrence of some higher concentrations. In many applications, therefore, the model must be capable of predicting the temporal frequency distribution at any given location. For example,

dispersion models can be used to compute seasonal or annual mean values, or medians with reasonable accuracy, but the accuracy may deteriorate at the higher end of the frequency distribution and therefore the prediction of the higher percentiles will be less reliable. In making these projections it is commonly assumed that the recent climatological/meteorological conditions will prevail in the future. However, these conditions may very well exclude extreme weather conditions which may occur occasionally (e.g. once every 5 or 10 years) and which can result in exceptionally high concentrations of pollutants for periods of several days. The manager of the AQMS may need to take such events into account when projecting air quality and when selecting the control strategy.

When real time control of ambient air quality has been selected as part of the control strategy (i.e. load reduction, load shifting or fuel switching), it may be necessary to have a model with the capability to predict concentrations on an hour-by-hour basis. In selecting the model it is important to keep in mind the type of results required for comparison with air quality objectives or standards and the acceptable limits of spatial and temporal resolution. These requirements will govern, or may themselves be governed by ambient or source monitoring, emission and meteorological data availability or the extent to which validation of the model has been successful.

As mentioned earlier, impact assessment is a potential instrument for integrated planning but has inherent difficulties due to a lack of baseline data and appropriate methodologies. Interests are conflicting and short-term and long-term considerations sometimes are divergent. The usefulness of modeling is important. However care must be taken in using very elaborate techniques when they depend on individual subjective assumptions.

Simple standardized assessment methods have to be recommended. Impact reports can play a real and valuable role in environmental management.

They encourage an integrated approach, foster public participation and may prevent decisions being made without full knowledge of the environmental impact.

Within the framework of an AQMS an impact report should contain information about the local situation, the focus of implementation and the means for implementing the proposal, the environmental consequences which are to be expected, the alternatives, a cost benefit analysis, the link with existing policy and a list of the sources from which the information was obtained.

Air quality impact statements can be defined as public documents, intended for decision making, in which the impact of proposed activities on air quality in the near and perhaps the more distant future is described. The physical system (the whole of natural environment, artifacts and localized activities) is in constant interaction with other ecological, economical and social systems.

IV.3.4. Selection of Control Strategy

Selection of control strategy is a step-by-step optimizing process. It consists primarily of the following steps:

1. Assessment
2. Apportionment of ambient air quality to sources
3. Identification of available control techniques
4. Integration of control techniques in alternative strategy plans
5. Selection of "best" plan

Each of these steps is discussed in detail below:

Step 1: Assessment

Assessment includes the collection, handling, evaluation, analysis and presentation of data necessary to understand the air pollution problem of the area and its causes. These data normally refer to geography, topography, land use, sources and emissions, ambient air quality, meteorology, climatology, atmospheric chemistry, etc. Other relevant data are population density and distribution, traffic patterns, energy production and consumption patterns.

Step 2: Apportionment of ambient air quality to sources

The concentration of a certain pollutant measured at a receptor point is normally caused by a variety of different sources or source categories. It is essential to identify their relative contribution to air pollution at receptor points. For example, it is important to know for a certain receptor point within the area of concern the percentage of ambient SO_2 concentration contributed by power stations, heating devices for residential and commercial buildings, diesel engines, chemical and metallurgical processes or refineries.

Model calculations may assist the accomplishment of this task. Using the data of the projected emissions inventory, apportionment must also be done for the future.

The result of apportionment is a scale, which ranks sources and source categories with respect to their contribution to air pollution within the area. This scale gives an idea of the sources whose emissions reduction would lead to the most effective reduction in air pollution.

Step 3: Identification of available control techniques

According to the scale mentioned before, for each source, all available techniques to reduce emissions have to be identified.

Step 4: Integration of control techniques in alternative strategy plans

Available control techniques for the relevant sources have to be combined in a strategy plan. Normally several alternative strategy plans are possible, all capable of meeting the goals of the AQMS. Each alternative has specific characteristics with respect to time scale, costs, socio-economic consequences and interaction with other environmental media, which must be specified as precisely as possible.

Step 5: Selection of "best" plan

The above alternatives are the basis for the authority to select the "best" strategy plan. Which plan is considered "best" depends on the selection criteria applied by the authority. Important selection criteria normally are the time required to meet the AQMS goals, the costs and socio-economic consequences.

IV.3.5. Surveillance

The establishment of emission standards as a part of the strategy plan requires the construction of a surveillance (monitoring) system to ensure compliance with standards. The same relates to ambient air quality (Fig. 3.5.-1).

Source Monitoring

Source monitoring may be done continuously, periodically or on a random basis. The monitoring applied to a certain source or source category depends on the amount and the substances emitted and on the variation of emissions with time. Standard methods for measuring stack emissions should be established for the enforcement of emission standards. The AQMS should, therefore, adopt a common reference method beforehand and prevent the proliferation of other measuring techniques that invariably yield greatly different results. Technical research may later justify the acceptance of alternate instrumentation.

The AQMS should select one method for emissions measurement and use it as the single technique for enforcing the standards as in the past. Source sampling test methods have been extensively used to determine whether sources are in compliance with emission standards. This is far from ideal, since the intermittent nature of sampling reveals little about emission variations as a function of process changes.

Continuous monitoring is the most appropriate method of surveying the emissions of major sources, plant operation, operation of gas cleaning devices and composition of fuels and raw materials. In cases where no continuous instrumentation is available, monitoring has to be done periodically or on a random basis with an appropriate sampling frequency, e.g. one sample per day. For monitoring of smaller stationary sources it would be sufficient to measure emissions periodically, e.g. once a year. Central heating burners and motor vehicles should also be included in this surveillance program because of the great number of these low level sources.

If source monitoring shows that emission standards are exceeded, appropriate emission reduction measures must be initiated immediately.

Data analysis and presentation must be closely related to the objectives of surveillance. This means that:

- data analysis has to produce the information which meets the specific information requirements of surveillance and

- data must be presented in a manner which is easily usable.

Most emission standards specify a single limit for emissions; those based on continuous monitoring will probably have a frequency distribution allowance similar to that used for ambient air standards. These data, in turn, lead the way to more advanced standards which will automatically allow for normal process variations yet enable the AQMS to continually oversee emission rates and take immediate corrective action when necessary.

The main objetive of data analysis is to condense numerous measured values for a defined period to characteristic values which may be related to standards.

These characteristic values may be
- the maximum value,
- certain percentiles of the cumulative frequency distribution,
- the arithmetic mean value,
- the frequency of exceeding the standard.

Depending on standard definition, these values may represent concentrations or mass flow rates within a defined time period (e.g. kg SO_2 per hour).

Ambient Air Quality Monitoring

Often the measurement system used for the determination of the existing ambient air quality is also designated to perform the surveillance in the AQMS. One has to keep in mind, however, that in selecting a specific control strategy, once the existing conditions concerning sources, emissions and ambient air quality are known, the questions to be answered by the surveillance may not be adequately covered by the measurement system that was originally established. Unless an extensive, and often sophisticated measurement system was used from the start, this difficulty often occurs. At that stage in practice it is difficult to rearrange or extend the existing facilities because of the financial investments involved.

The surveillance can be continuous or periodical. It can serve different purposes, e.g. act as an early warning system to alert when high levels or standards are being approached or exceeded; determine the trend with time (from year to year); determine the transmission (transport, deposition, import and export from and to countries) or give a description of the geographical distribution and occurrence of the pollutants.

In most surveillance systems, especially the more detailed ones that also cover regional and local problem areas, mobile units are used in addition to the fixed reference measurement stations. The advantage of the mobile units is, in this case, that they can cover a large area within a relative short period of time. When compliance with ambient air quality standards has to be determined the representativeness of the measurements for a larger area has to be considered. Any measuring equipment should be able to determine a level of a pollutant as specified by a standard or the criteria with an accuracy of at least ten percent of that level. An improvement can be gained when suitable sensing equipment is used. With these instruments the space and time dependencies can be more adequately determined. Provisions have been made within the AQMS that the surveillance system can be revised as soon as it becomes clear that it does not meet the objectives as laid down in the strategy plan.

Important points are the requirements for updating of the mesaurement equipment and for ensuring that the measurement methods are equivalent and comparable to an acceptable reference procedure, for ensuring that the equipment is properly maintained and for providing for training and operating technicians. In summary, there is a need for thoroughly planned and continuously operated quality assurance programs.

The data analysis and presentation methods to be used are, in most cases, comparable to the ones needed for the analysis of the existing ambient air quality. A difference, however, is the data presentation used to answer the specific question asked in the AQMS. Another problem is whether one should store and keep ready for retrieval the reduced data or the original measurement data. One has to keep in mind that for purposes other than the direct questions asked by the surveillance requirements, there may be a need for a renewed study or research based on the original measured data. Especially for trend analysis and studies concerning the transmission of air pollutants, it often becomes necessary, when new analytical techniques for data analysis become available, to formulate new criteria on which the data

analysis should be based. In this case a new set of reduced data is required. For continuous measurements, one should aim to store only the validated measurement data if retrieval for future use is anticipated.

IV.3.6. Interaction with other Environmental Media

The complexity of the problems involved in the protection of the environment from pollution should not be underestimated. Even the slightest pollution may lead - like all human activities - to changes in environmental systems. To understand these changes it is necessary to know the sources, concentrations, and properties of pollutants, the pathways they take through the environment and the nature of their interactions with other pollutants, receptors and sinks. At any stage or intersection in these pathways, pollution may have significant social, economic, ecological or toxicological implications for people and their life support systems. These implications will vary greatly according to the nature and source of the particular pollution process, the extent to which resulting pollutants are transformed at various stages in environmental pathways, and the sensitivity of the receptors. It is very difficult, therefore, to isolate particular pollution problems for meaningful control strategy. Consequently the environmental media (air, water and soil) should not be considered independently; they are linked together with many direct and indirect connections.

Air Quality Management Systems therefore can never be seen isolated from other environmental management programs. The programs for the protection of the other environmental media must be carefully balanced with the air pollution strategy plan to ensure that an improvement in air quality will not lead to deterioration in other media.

Examples of the direct interaction of air pollutants with other media are:

- sulphur dioxide and nitrogen oxides can be absorbed in atmospheric water droplets and cause increased acidity of rain (and therefore in the soil) in nearby and remote areas,

- power stations need direct cooling water systems or cooling towers and the waste heat will either be released into rivers or into the atmosphere,

- certain heavy metals may affect vegetation, thus introducing them into the food chain,

- the emissions of dust mostly from coal-fired furnaces or metallurgical plants which contain certain heavy metals, lead to sedimentation of dust containing these heavy metals.

One example of indirect interaction (caused by air pollution abatement measures) is:

- gas cleaning devices very often use wet scrubbing systems to absorb emitted substances; the scrubbing water will sometimes be released into rivers and the pollutants may be introduced into the food chain; similar problems may arise if the precipitated substances or their reaction products are deposited.

The fates of air pollutants and their reaction products are different after they have been released into the atmosphere.

IV.3.7. Revision of Strategy Plans

Normally a plan will not work from the beginning to the end without any revision. It is therefore necessary to check its success and to keep it oriented towards the original or revised objectives. Because a strategy plan is designed for a time scale of about 10 or 20 years, a

revision of strategy plans must be an essential part of AQMS. In general, revision is necessary if the plan is inadequate to attain or maintain the adopted standards or objectives.

Also, a review of the plan for possible revision should be made after fixed periods, e.g. after every 3 to 5 years. These reviews will guarantee that assumptions and boundary conditions made in plan design are checked and kept up to date. Independent of these periodic reviews the plan must be revised if something substantial happens to affect the plan or its objectives. Plan revision is necessary, if:

- the legal basis for air pollution control has changed,
- new emission, product or air quality standards are set,
- substantial changes take place in the emissions,
- new scientific findings about nature and extent of effects are available,
- new control technology becomes available,
- an unexpected long-term shortage of a certain fuel occurs,
- a new energy supply system is established, e.g. using the waste heat of a large power plant for space heating.

IV. 4. Main Sources of Air Pollution

The main sources of air pollution have long since been identified as:

- power plants,
- transport,
- industry,
- households.

However, it is still difficult to give exact figures on the emissions of each of these sources and the main pollutants. Data which were due by the middle of 1985, compiled by an international EC study group, are unfortunately still not available.

Therefore the authors of this study reviewed data published by OECD, EC and national authorities in order to get reliable estimates of the SO_2-, NO_x-, HC- and particulate emissions.

The results of the review, expressed as a rough split into source categories, are displayed in Table IV.3.-1. to IV.3.-3.

For HC and particulates no breakdown of data into national emissions was possible. Therefore only EC-wide estimates are shown in Table IV.3.-3.

In spite of the limited knowledge on emissions, the data shown allow some general conclusions:

1. The main sources of the pollutants under consideration in this study are clearly identified:

   for SO_2: power plants and industries,
   for NO_x: transport and power plants,
   for HC : transport and use of volatile organics for particulate industries and power generation.

Moreover it is evident that these emission structures are quite similar in all Member States, despite the fact that some occur between one country and another.

2. Overall reduction measures must also take into account the natural contributions to total emissions. They are estimated to be:

   - 10% of total emissions for SO_2,
   - 20% of total emissions for NO_x,
   - 50% of total emissions for HC.

   Little is known about the relevance of natural particulate emissions in Europe; therefore they are not taken into account.

3. If a global reduction of total emissions within the EC is required, four Member States should make special efforts: France, the Federal Republic of Germany, Italy and the United Kingdom, because these Member States alone are responsible for:

   - 86% of total man-made SO_2 - emissions, and
   - 85% of total man-made NO_x - emissions.

IV.5. Proposals for Elements of a Strategy

IV.5.1. Introduction

The complexity of the issue, as demonstrated in the previous chapters, does not allow the proposition of one single overall strategy. It is evident that specific approaches have to be developed for each pollutant and/or source category.

Moreover it seems, in some cases, to be worthwhile to differentiate between local and regional problems: pollution by suspended particulates is obviously a local problem, while pollution by SO_2, NO_x and O_3 is also a European-wide problem.

Things become even more complicated because the knowledge on effects of pollutants at low concentration is incomplete. The comprehensive review given in Chapter II on effects and investigations on measured ambient air levels showed that effects can be explained only for urban areas. However, there is a strong believe that air pollution is also responsible for effects in rural areas, e.g. forest decline.

It is difficult to recommend a straightforward policy in such an unclear situation. In particular the "immission-standard" approach, as outlined in all its principles in Chapter IV.2, cannot be applied nationwide any more because:

- clear "non-adverse effect levels" which guarantee the protection human health and, in particular, the environment, cannot be defined;

- even if they could be defined, it would be very difficult to identify the single sources responsible for a certain pollution in a particular rural area;

- adequate monitoring, a necessary prerequisite for any "immission-standard" approach is difficult to perform in rural areas.

The only appropriate policy is therefore

i. to reduce the relevant emissions in urban areas down to those levels identified as "non-adverse effect levels";

ii. to introduce a long-term policy of an overall reduction of emissions from all sources important for long-range transport of pollutants.

With regard to point i., the European Commission has already taken the necessary steps in fixing ambient air limit values for SO_2, particulate matter, NO_2 and lead and accompanying these measures by some concrete

proposals to reduce emission from important sources. This approach should be reinforced by setting out supplementary emission standards for important sources or adequate product norms, whenever necessary. However, because the air pollution problems differ from one urban area to another, the European Commission should leave as much flexibility as possible to local authorities and check only regularly the success of the measures taken. Emission standards and product norms should be set out only if the harmonization of the market or the safeguarding of fair competition requires such measures.

In this context, the measures taken, proposed by the European Commission, on lead in petrol, sulphur content of gasoil and in particular emissions from passenger cars are most helpful. Some of these measures can be considered at the same time as a contribution to the long-term policy of point ii.

With regard to point ii., the Commission should apply the AQMS-instruments

- emission standards and
- product norms,

supplemented by an active financial support for reduction measures, in those Member States where the economic situation is difficult, and comprehensive research and development work on clean technologies.

Source elements for such approaches are outlined below for SOH_2, NO_x, particulates and ozone. Hydrocarbons are not included here because they were studied in this paper only in their role as precursors of ozone.

IV.5.2. Strategies to reduce SO_2

As shown in Table IV.3.1, three sources substantially contribute to SO_2-pollution;

- power plants,
- industries,
- households.

The Commission attacked emissions from large power-plants and large industrial combustion installations by its proposal COM(83) 704 and emissions from households partly by its proposal COM(85) 377. Both proposals have some shortcomings.

i. The proposal on large combustion installations does not include any financial support system and/or derogations for "poor" Member States and is therefore unacceptable to some Member States.

ii. The proposal on the S-content of gasoil gives a lower limit value which is unnecessary and which makes local policies more difficult.

However, both approaches point into the right direction and would reduce total SO_2-emission, if adopted, by approximately 37%, equal to 5800 kilotonnes. Supplementary measures are possible and necessary:

i. the setting out of limit values for industrial sources,
ii. the limiting of the S-content in coal and heavy oils.

With regard to point i, emission standards should be laid down for all combustion installations larger than 5 MW therm. The standards should be in such a range that washed coal (0,8% - 1%S) and low sulphur oil (1% - 1,2%S) is allowed to be burnt with no further post-treatment of the exhaust gases necessary in order to comply with the limit values. With regard to point ii, it should be forbidden to burn oil with more than 0,3% S and coal with more than 0.8% S in domestic installations. Moreover, the Commission should step up its activities on energy savings and research and development work on clean technologies for industry.

It can be estimated that such measures would cut down SO_2 - emissions by 40% from industrial sources and by 30% from domestic sources, resulting in a supplementary reduction of approximately 2000 kilotonnes, equal to 13% of actual SO_2 emissions. Together with the already proposed measures, an overall reduction of 50% would be achieved.

Little can be said at present about the costs involved with such measures. However, as estimates, the data given in Figure IV.4.2.1 may be helpful. They demonstrate that averaged costs in the range of 250 to 2000 US$ per ton SO_2 removed, depending on the strategy chosen, can be expected. That means in more detail that

i. the implementation of the European Commission's proposal on large combustion installations would cost the Member States approximately 3 to 6 billion US$ per year;

ii. the implementation of low sulphur gasoil to an averaged EC-level of 0.2% S would cost the Member States approximately 300 to 400 million US$ per year;

iii. the implementation of emission limit values for industrial sources would cost Member States approximately 200 to 400 million US$ per year;

iv. the implementation of low sulphur coal and oil for domestic heating would cost Member States approximately 400 to 600 million US$ per year.

However, it should be kept in mind that these figures are nothing more than broad estimates, indicating the order of magnitude of the costs.

Finally, it is absolutely necessary to accompany these measures by political action on the ECE-level because the Member States of the EC are only responsible for approximately 25% of total European SO_2-emissions. Any further reductions in the EC Member States would be small in comparison to the emission of others, in particular East-European countries.

IV.5.3. Strategies to reduce NO_x

Three main sources of man-made NO_x-emissions have been identified and listed in Table IV.3.2:

- transport,
- power plants,
- industry.

The European Commission has submitted to Member States two main proposals with a view to reduce emissions from two sources:

- emissions from passenger cars will, most likely, be reduced due to the Council agreement of June 1985, based on the Commission's proposal COM(85) 288,

- emissions from large combustion installations are attacked by proposal COM(83) 704, which aims at a 40% overall reduction of emissions from this source.

The implementation of these proposals would result in an overall reduction of NO_x-emissions of approximately 3000 kilotonnes by 1995, equal to 29% of total man-made emissions.

As in the case of SO_2, supplementary measures are possible, which may result in further substantial reductions:

i. the laying down of limit values for motor vehicles above 3.5 tonnes weight,

ii. the laying down of emission standards for combustion installations in the range of 5 MW and 50 MW_{therm}, including industrial boilers.

Such measures would reduce NO_x-emission by, approximately, another 900 kilotonnes, equal to 9% of total man-made emissions, so that an overall reduction of 38% seems to be achievable.

Measures, as proposed in point i., are underway and the Commission will put forward appropriate proposals by the end of 1985.

Proposals concerning point ii. are under consideration and studied by working groups of the Commission.

The available information on emission structure in Member States and the measures planned is so limited, as yet, that no sound estimate on costs can be given.

As in the case of SO_2, it recommended to enforce activities on the ECE-level because a substantial part of the total man-made NO_x is emitted from within non-Member States of the EC. An efficient European overall reduction strategy requires that all European countries reduce emissions.

IV.5.4. Strategies to reduce Particulates

Only very rough estimates on EC-wide emissions of particulate matter are available. It can be concluded from these estimates that two sources are of major importance:

- industries
- power plants.

With regard to power plants, a proposal has already been made by the European Commission (COM(83) 704)) which would, if adopted, result in a 40% reduction of emissions from this source, equal to approx. 9% reduction of overall emissions.

The main source of particulate emissions, industries, is however still not covered by the Commission's activities and it is highly recommended to work out appropriate proposals. It is extremely difficult, due to the lack of data, to estimate the reduction potential of such proposals because emissions have already substantially been reduced in the last decade by some Member States; others however have done little. Nevertheless, a 30% reduction of emissions from industrial sources seems to be still possible on an EC-wide level, resulting in a 17% reduction of overall emissions. As in the case of SO_2 and NO_x, the best way to achieve such reduction seems to be by setting out emission standards for the various industries. Old installations should be obliged to meet the standard within an appropriate delay.

IV.5.5. Strategies for Limiting Oxidants

Since oxidants are secondary air pollutants, the only way to limit them is to curb emissions of their precursors. The first step must be to establish the quantitative relationship between the imission concentrations for the oxidants and those for their precursors, and then between the imission and emission concentrations for the precursors. Any measures to curb emissions of hydrocarbons (HC) and/or oxides of nitrogen (NO_x) must be based on figures for such relationships.

Simulation exercises and model calculations have yet to produce any recommendations of a quantitative nature - in other words, indicating the percentage by which maximum ozone levels would fall if emissions of one or both precursors (i.e. of HC or of NO_x) were reduced by a set amount.

Nevertheless a large number of estimates which could serve as a basis for making recommendations have been published:

i. Proportionality between the maximum ozone level and hydrocarbon emissions (total NMHC)

The degree of proportionality can be determined empirically. However, only in highly polluted conurbations in California has this proved a practical method of reducing emissions, but not in the outskirts of towns and cities or in rural areas.

ii. Statistical relationship between the maximum ozone level and precursor concentrations or meteorological factors

Non-linear correlation, based on hydrocarbon readings measured in the open air between 06.00 and 09.00 hours and on the daily maximum ozone level, between a reduction in hydrocarbons alone by a given percentage and the highest maximum ozone level measured in the urban area concerned.

Although this method can be used in areas where the readings needed to establish the correlation have been taken, it brings about a greater reduction in hydrocarbon levels than method i.

Another statistical method for predicting ozone concentrations is based on the correlation between such meteorological factors as the maximum daily temperature, (solar) radiation and windspeed on the one hand and maximum ozone levels on the other. However, forecasts based on correlations measured in this way are valid only for the measurement site in question and need data covering many years.

Statistical models for six-hour to 30-hour episodes have also been used to forecast the maximum average hourly ozone value. In this case, a correlation is established between the ozone data on the one hand and the temperature at 850 mbars plus the altitude and thickness of the blocking layer on the other.

iii. Relationship between maximum ozone level and concentrations of hydrocarbons and of oxides of nitrogen

This is an isopleth model based on NMHC and NO_x imission concentrations between 06.00 and 09.00 hours and on the one-hour mean maximum ozone level in the afternoon. Taking an analytical approach to the EKMA method, a linear relationship, within certain concentration ranges, between the maximum ozone value on the one hand and the geometric mean of the precursor values on the other was established in 1982 in the USA.

Isopleths (lines showing identical maximum ozone levels) for different concentrations of precursors have been plotted from smog chamber experiments and calculations with chemical and kinetic models, such as the EKMA method. But each of the isopleths plotted so far points to a very different sequence of events.

Because of the enormous volume of data required, very few comparisons with outside-air measurements have been made.

Therefore it can be concluded that, at the moment, there are too many unanswered questions about the abatement strategies suggested by the different forecasting models to make any specific recommendations. Nevertheless, it is fair to assert that any kind of reduction in precursor emissions will reduce the average total pollutant load over wide areas, even if it fails to produce any significant reduction in the ozone load locally.

To provide a solid basis for decisions to reduce oxidant levels, first physical and chemical dispersal models must be further developed. This, however, will entail expanding the meteorological station networks and refining the emission data to make them less sensitive to annual or daily events or to the procedure used. Secondly, continuous-monitoring stations should be set up in areas where oxidants present a

particularly high risk. This could cast light on the relationship between precursor concentrations and oxidant formation and also on how they are affected by meteorological conditions. Once the relationships have been substantiated in this way, they can provide a basis for recommending steps to reduce levels in the area where the measurements were taken.

On the basis of the measurements published so far, it is recommended that instead of running large measurement networks to record ozone levels over wide areas, ozone concentrations should be recorded continuously at selected reference sites, from which airborne measurements could than be made at different altitudes and in different weather conditions to determine how the photochemical oxidants are dispersed over larger areas and to pinpoint the sources of precursors.

Regional measures to reduce emissions are unlikely to be enough in themselves to keep ozone levels within non-effect limits. On the other hand, regional measures of this type could probably prevent regional or local peaks. Consequently, a coordinated campaign to reduce emissions throughout the whole of Europe appears the only way to reduce the ozone load over wide areas.

At this stage it is still too early to give any generally valid figures for the reduction in oxidant levels which can be expected from any given measure to limit emissions of individual categories of precursor. The limited, yet informative, airborne measurements made in various conurbations in the Federal Republic of Germany have shown that high oxidant concentrations are particularly common downwind from hydrocarbon sources. Consequently, imposing limits on hydrocarbon emissions would seem to be one promising way of curbing high ozone loads attributable to sources in the region concerned itself. Above all, emissions of hydrocarbon components which are found in medium or high concentrations and are relatively quick to react with hydroxyl radicals in the atmosphere must be reduced. At supra-regional and European level, reducing NO_x levels presumably takes precedence over

reducing hydrocarbons since, on a much larger scale, reactive hydrocarbons from natural sources also play their part in the formation of oxidants. Before reducing hydrocarbon emissions alone, one must first carefully check that this would not simply shift the oxidant load from the regional to the supra-regional level.

Since weather also affects oxidant formation, the oxidant load fluctuates more from year to year than primary immissions of NO_x or hydrocarbons. This fluctuation must be taken into account in the abatement measures to ensure that the limit value laid down is never exceeded, not even under unfavourable meteorological conditions.

TABLES

Table I.2-1: Examples of ambient air quality standards for SO_2

Country	Average time Statistical definition		concentration in ug/m³	Remarks
China[9]	Annual daily average	class I	20	
		class II	60	
		class III	100	
	Daily average	class I	50	
		class II	150	
		class III	250	
	Maximum at any time	class I	150	
		class II	500	
		class III	700	
Canada[3]	Annual arith. mean		60	
	24 h mean		300	
	1 h mean		900	
Belgium, France, Greece, Ireland, Luxembourg, The Netherlands, United Kingdom[1]	Annual median of 24 h values		80 if Black Smoke > 40 120 if Black Smoke ≤ 40	
	Winter median of 24 h values		130 if Black Smoke > 60 180 if Black Smoke ≤ 60	
	Annual 98-percentile of 24 h values		250 if Black Smoke >150 350 if Black Smoke ≤150	

cont'd.

Country	Average time Statistical definition	concentration in ug/m^3	Remarks
F.R. Germany, Denmark, Italy[2]	Annual arith. mean of 1/2 h values	140	
	Annual 95-percentile of 1/2 h values	400	
Japan	24 h average of 1 h means	106[4]	
	1 h mean	266[4]	
Spain	Max. 1/2 h mean	700	
	Max. 24 h mean	400	
	Max. monthly arith. mean	250	
	Max. annual arith. mean	150	
Sweden	Winter arith. mean/max. value which should not be exceeded	100	
	Annual 24 h mean	300	
	Annual 1 h mean/should not be exceeded for more than 1 percent of the time per month	750	
Switzerland[8]	Annual arith. mean of 1/2 h values	30	
	95-percentile of 1/2 h values	120	

Table I.2-3: Ambient air quality standards for particulates, measured as black smoke in a selected number of industrialised countries

Country	Average time Statistical definition	concentration in ug/m^3	Remarks
Belgium, France, Greece, Ireland, Luxemburg, The Netherlands, United Kingdom	Annual median of daily means	80	
	Winter median of daily means	130	
	Annual 98-percentile of daily means	250	
Sweden	Winter arith. mean, should not exceeded	40	
	24h-mean, should not be exceeded for more than 2 days	120	

cont'd.

Country	Average time Statistical definition	concentration in ug/m³		Remarks
Switzerland[7]	Annual arith. mean of 24 h values	70[5]		
	95-percentile of 24 h values	150[5]		
United States	Annual geometric mean	75[2,5]	60[3,6]	
	24 h mean, not to be exceeded once per year	260[2,5]	150[3,6]	

(1) Not mandatory, but proposals which can be incorporated in provincial laws

(2) Total suspended particulates

(3) Particulates 10 cm in diameter

(4) In application of Directive 80/779/EEC

(5) Primary standard

(6) Secondary standard

(7) Proposal of May 1984

(8) Definition of classes, see Table II.2-1

Table I.2-4: Ambient air quality standards for O_3 in a selected number of industrialised countries

Country	Average time Statistical definition		concentration in ug/m³	Remarks
Canada[1]	Annual arith. mean		30	reference temp. 25°C
	24 h - mean		50	reference pressure
	1 h - mean		160	760 mm mercury
China[2]	Hourly average	class I	120	
		class II	160	
		class III	200	
Japan	1 h mean		119	
Switzerland[4]	95-percentile of 1/2 h values		80	
United States	1 h mean/not to be exceeded more than one time per year		240[3]	reference temp. 25° C reference pressure 760 mm mercury

(1) Not mandatory, but proposals which can be incorporated in provincial laws

(2) Definition of classes see Table II.2.-1

(3) Primary standard

(4) Proposal of May 1984

Table I.2-5: Ambient air quality standards for NO_2 in a selected number of industrialised countries

Country	Average time Statistical definition	concentration in ug/m³	Remarks
Canada[1]	Annual arith. mean	100	reference temp. 20°C
	24 h - mean	200	reference pressure
	1 h - mean	400	760 mm mercury
F.R. Germany	Annual arith. mean of 1/2 h values	80	
	98-percentile of 1/2 h values	300[2]	
Japan	1 h - mean, daily average of hourly values	76 - 115	
The Netherlands[3]	Median of daily mean, 1 h values	50	
	98-percentile, daily mean values	120	
	98-percentile, 1 h mean values	135	
Switzerland[6]	Annual average of 1/2 h values	40	
	95 percentile of 1/2 h values	120	
United States	Annual arith. mean	100[4]	
USSR	24 h value (not to be exceeded)	85[5]	
	1/2 h value (not to be exceeded)	85[5]	

(1) Not mandatory but proposals which can be incorporated in provincial laws

(2) Will be replaced by 1070 1987 by 200 ug/m³, calculated as a 98-percentile of 1 h values measured throughout the year (Directive 85/203/EEC)

(3) Interim guidelines

(4) Primary standard

(5) In USSR values are combined, see text

(6) Proposal of May 1984

Table I.2-6: Ambient air quality standards for non-methan hydrocarbons in a number of industrialised countries

Country	Average time Statistical definition	concentration in ug/m^3	Remarks
Spain(1)	Max. 1/2 hr - mean	280	calculated as $C_6 H_{14}$
	Max. 24 hr - mean	140	
Japan(2)	3 hr average of 1 hr - mean	132 - 205	calculated as CH_4
United States	3 hr - mean	160	calculated as CH_4

(1) Mandatory ambient air quality standards

(2) Given not as a standard but as a guideline

Table I.3-1: Emergency levels as applied in a number of industrialised countries

(values in ug/m³, if not indicated differently)

Country		SO_2	CO	NO_2	non-methan HC	parti culates	O_3	combined values	Remarks
F.R. Germany[1]	stage 1	800	30000	600	5000	none	none	none	At least one of these concentrations has to be excluded for at least 3 hrs
	stage 2	1600	60000	1200	10000				
	stage 3	2400	90000	1800	15000				
F.R. Germany[2]	stage 1	600	30000	600	none	none	none	11000[3]	At least one of these concentrations has to be excluded for at least 3 hrs
	stage 2	1200	45000	1000				14000[3]	
	stage 3	1800	60000	1400				17000/ 14000[3]	
Greece	stage 1	200[4]	200[5]	15000[6]	none	3[7]	none	none	
	stage 2	400[4]	500[5]	25000[6]		5[7]			
	stage 3	500[4]	700[5]	35000[6]		7[7]			
Japan	stage 1	532[8] 798[9]	none	none	none	2000[11]	239[10]	none	Conversion from ppm to ug/m³ at 20°C and 760 mm mercury
	stage 2	1330[8] 1862[9]				3000[11]	796[10]		
Spain	stage 1	800	34000	565	none	600[13]	none	327000[13,14]	All values as 24h-averages
	stage 2	1400	46000	750		1000[13]		600000[13,14]	
	stage 3	2200	60000	1000		1600[13]		1000000[13,14]	
United States		2660	57500[17] 86300[18] 144000[16]	938[15] 3750[16]	none	1000[13]	1200	none	
California (USA)	stage 1	13150[20] 526[21]	46000[20] 23000[22]	none	none	none	400		converted from ppm into ug/m³ for 25°C and 760 mm mercury
	stage 2	26300[20] 1841[21]	86250[20] 40250[22]				700		
	stage 3	52600[20] 2367[21]	115000[20] 57500[22]				1000		

cont'd.

(1) As applied in Bayern, Hessen, Baden-Wuerttemberg, Saarland and Rheinland-Pfalz
(2) As applied in Nordrhein-Westfalen and most likely by others in the near future
(3) Combined concentration of SO2 + 2 x particulates
(4) 24h-average
(5) 1h-average
(6) 8h-average, calculated from 1h-mean
(7) Coefficient of haze, 24h-averages, calculated from 3h-mean
(8) 3h-average, calculated from 1h-mean
(9) 2h-average, calculated from 1h-mean
(10) 1h-mean
(11) Particulates smaller than 10 um, 2h-average, calculated from 1h-mean
(12) Particulates smaller than 10 um, 3h-average, calculated from 1h-mean
(13) Total suspended particulates
(14) Combined effect SO2 particulates
(15) 24h-mean
(16) 1h-mean
(17) 8h-mean
(18) 4h-mean
(19) 24h-mean SO2 * 24h-mean TSP
(29) 1h-mean
(21) 24h-mean
(22) 12h-mean
(23) These levels shall apply when the oxidant concentration and the sulphur dioxide concentration exceed 0.10 ppm and shall be determined by adding the oxidant and sulphur dioxyde concentrations

TABLE II.1.-1 EFFECTS OF EXPOSURE TO POLLUTANT MIXTURES ON PULMONARY FUNCTION IN CONTROLLED STUDIES OF HEALTHY HUMANS

Pollutant	Concentration ug/m^3	No. of subjects	Exposure time (minutes)	Effects	Reference
SO_2 O_3	980 740	8	120	Decrease pulmonary functions (in synergistic effect of SO_2 on O_3) FRC, $FEV_{1.0}$, MMFR, $MMFR_{50\%}$	HAZUCHA and BATES, 1975 BATES and HAZUCHA, 1973
SO_2 O_3	980 740	4 (normal) 4 (ozone sensitive) 4 (from Bates)	120	Unable to confirm synergistic effects, pulmonary decrement due to O_3 alone	BELL ET AL., 1977
SO_2 O_3	1060 800	9	120	Unable to confirm synergistic effects, changes due to ozone alone	HORVATH and FOLINSBEE, 1977 BEDI ET AL., 1979
SO_2 O_3	400 300	6	120	Significant enhanced decrease in SG_{aw} after exposure to SO_2 + O_3 in comparison to O_3 alone	KAGAWA ET AL., 1979
O_3 NO_2	980 940	8	30	No additive effect or interaction between ozone and nitrogen dioxide was observed under different environmental conditions	HORVATH ET AL., 1979
NO_2 SO_2	9550 13300	11	120	No changes in PaO_2, $PaCO_2$, TGV · R_{aw} increased	VON NIEDING ET AL., 1979

TABLE II.1.-1 EFFECTS OF EXPOSURE TO POLLUTANT MIXTURES ON PULMONARY FUNCTION IN CONTROLLED STUDIES OF HEALTHY HUMANS

cont'd 1

Pollutant	Concentration ug/m^3	No. of subjects	Exposure time (minutes)	Effects	Reference
NO_2 SO_2	940 1330	24	120	No changes in pulmonary functions	LINN ET AL., 1980
NO_2 SO_2	940 810	19 (asthmatics)			
SO_2 NaCl	4256 - 13300 (0.22 um MMD)	13	5	Synergistic increases in airway resistance with aerosol	TOYAMA, 1962
SO_2 NaCl	23940 - 159600 (MMD = 0.95 um)	10	5	Airway resistance greater after exposure to aerosol than to exposure to SO_2 alone	NAKAMURA, 1964
SO_2 Saline particles	1330 - 13300 (7.0 um)	9	15	$MEF_{50\%}$ significantly greater, decreases in aerosol (NaCl) condition	SNELL and LUCHSINGER, 1969
SO_2 NaCl	2660 1000 (MMD 0.9 um, Sg = 2.0 um)	8 (asthmatics)	30	$\dot{V}_{max\ 50\%}$, $\dot{V}_{max\ 75\%}$, $FEV_{1.0}$ and R_t decrease significantly in aerosol condition	KOENIG ET AL., 1981
SO_2 NaCl	3000 - 9600 2 - 2,7 (MMD 0.25 um)	10	30	No effect on pulmonary functions	BURTON ET AL., 1969

TABLE II.1.-1 EFFECTS OF EXPOSURE TO POLLUTANT MIXTURES ON PULMONARY FUNCTION IN CONTROLLED STUDIES OF HEALTHY HUMANS

cont'd 2

Pollutant	Concentration ug/m^3	No. of subjects	Exposure time (minutes)	Effects	Reference
SO_2 NaCl	2660 - 45220 10000 - 30000 (MMD 0.15 um)	12	30	Changes in pulmonary function similar to changes due to SO_2 alone, not influences by aerosol	FRANK ET AL., 1964
SO_2 NaCl	2660 1000 (MMD 0.9 um Sg = 2.0 um)	9 (asthmatics)	60	Significant decreases in $\dot{V}_{max50\%}$ and $\dot{V}_{max\ 75\%}$	KOENIG ET AL., 1980
SO_2 NaCl	2660 1000 (MMD 0.9 um Sg = 2.0 um)	(normals)	120	No pulmonary effects demonstrated	MORGAN ET AL., 1977
SO_2 O_3 H_2SO_4	980 740 100 (MMD 0.5 um, Sg = 3.0)	19 (normals)	120	Small but statistically significant decrements in FEV_1 and slight increases in the incidence of clinical symptoms	KLEINMAN ET AL., 1981

TABLE II.1.-1 EFFECTS OF EXPOSURE TO POLLUTANT MIXTURES ON PULMONARY FUNCTION IN CONTROLLED STUDIES OF HEALTHY HUMANS

cont'd 3

Pollutant	Concentration ug/m^3	No. of subjects	Exposure time (minutes)	Effects	Reference
O_3	1 000,	4	240	With each group minimal alterations in pulmonary function caused by O_3 exposure. Effects were not increased by addition of NO_2 or NO_2 and CO to test atmospheres.	HACKNEY ET AL., 1975
O_3 with NO_2	1 000 with 560				
O_3 with NO_2 and CO	1 000 with 560 and 45 000				
O_3	500,	7	120	Little or no change in pulmonary function found with O_3 alone. Addition of NO_2 or of NO_2 and CO did not noticeably increase the effect. Seven subjects including some believed to be unusually reactive to respiratory irritants	HACKNEY ET AL., 1975
O_3 with NO_2	500 with 560				
O_3 with NO_2 and CO	500 with 560 and 45 000				
SO_2 with NO_2 and O_3	13 300 with 9 550 and 200	11	120	No changes in PaO_2, $PaCO_2$, $TGV \cdot R_{aw}$ increased	VON NIEDING ET AL., 1979
SO_2 with NO_2 and O_3	320 with 115 and 50	11	120	No changes in pulmonary functions	VON NIEDING ET AL., 1979

TABLE II.1.-1 EFFECTS OF EXPQSURE TO POLLUTANT MIXTURES ON PULMONARY FUNCTION IN CONTROLLED STUDIES OF HEALTHY HUMANS

cont'd 4

Pollutant	Concentration ug/m^3	No. of subjects	Exposure time (minutes)	Effects	Reference
SO_2 NO_2 O_3	810 280 140	15	8 hr/day for 4 successive days	No changes in pulmonary functions	ISLAM and ULMER, 1979
NO_2 O_3 SO_2	100 with 50 and 300	11	120	No effect on R_{aw} or $AaDO_2$; exposed subjects showed increased sensitivity of bronchial tree to a bronchoconstrictor (acetylcholine) over controls not exposed to pollutants.	VON NIEDING ET AL., 1977

TABLE II.1.-2 QUANTITATIVE COMMUNITY HEALTH EPIDEMIOLOGICAL STUDIES ON EFFECTS OF EXPOSURE TO POLLUTANTS MIXTURES ON PULMONARY FUNCTION

Pollutants	Measure	Exposure concentrations ug/m^3	Study population	Effect	Reference
O_3 and others	lowest estimated effect level average maximum hourly concentration range	300 O_3 200 to 300 O_3	Adolescents	Increased airway resistance associated with ozone, sulfur dioxide, and temperature	KAGAWA and TOYAMA, 1975 KAGAWA ET AL., 1976 KAGAWA, 1983
O_3 and others	lowest estimated effect level average maximum hourly concentration range	160 O_3 200 to 240 O_3	Adult asthmatics	Decreased daily peak flows and increased prevalence rate for acute symptoms associated with ozone, low temperature, and high total suspended particulate matter.	LEBOWITZ ET AL., 1982, 1983 LEBOWITZ, 1984
O_3 and others	lowest estimated effect level average maximum hourly concentration range	160 O_3 200 to 240 O_3	Children and young adults	Daily peak flows decreased 12.2 to 14.8 % with ozone and total suspended particulate matter	LEBOWITZ ET AL., 1982, 1983 LEBOWITZ, 1984

TABLE II.1.-2 QUANTITATIVE COMMUNITY HEALTH EPIDEMIOLOGICAL STUDIES ON EFFECTS OF EXPOSURE TO POLLUTANTS MIXTURES ON PULMONARY FUNCTION

cont'd 1

Pollutants	Measure	Exposure concentrations ug/m^3	Study population	Effect	Reference
NO_2 and SO_2	Mean "annual" 24-hr concentrations: high exposure area low exposure area 1-hr mean: high exposure area low exposure area	103 NO_2 92 SO_2 75 NO_2 36 SO_2 260 to 560 NO_2 110 to 170 NO_2	Pulmonary function tests administered to 128 traffic policemen in urban Boston and to 140 patrol officers in nearby sub-urban areas.	No difference in various pulmonary function tests.	SPEIZER and FERRIS, 1973 BURGERS et AL., 1973
SO_2 and SPM	24-hr average	≥1000 BS ≥1000 SO_2	Persons with existing respiratory or cardiac disease	Likely increases in daily mortality above a 15-day moving average during winter 1958 - 1959.	MARTIN and BRADLEY, 1980
	24-hr average	750 - 1000 BS 710 - 1000 SO_2	Persons with existing respiratory or cardiac disease	Slight indications of likely increases in daily total mortality above a 15-day moving average during winters of 1958-59 and 1959-60	MARTIN 1964

TABLE II.1-2 QUANTITATIVE COMMUNITY HEALTH EPIDEMIOLOGICAL STUDIES ON EFFECTS OF EXPOSURE TO POLLUTANTS MIXTURES ON PULMONARY FUNCTION

cont'd 2

Pollutants	Measure	Exposure concentrations ug/m^3	Study population	Effect	Reference
	24-hr average	750 - 1000 BS 710 - 1000 SO_2	Persons with existing respiratory or cardiac disease	Slight indications of likely increases in daily total mortality above a 15-day moving average during winters of 1958-59 and 1959-60	MARTIN, 1964
	24-hr average	250 - 500 BS 500 - 600 SO_2	Group of chronic bronchitis patients	Likely worsening of health status	LAWTHER ET AL., 1958, 1970
	24-hr average	< 250 BS < 500 SO_2	Group of chronic bronchitis patients	No apparent response or worsening of health status	LAWTHER ET AL., 1970, 1975
	annual average	100 - 200 BS 150 - 200 SO_2	British postal workers	Greater prevalence of cough and phlegm in areas of elevated pollution	LAMBERT and REID, 1970
		230 - 300 BS 180 - 275 SO_2	Children	Likely increased frequency of lower respiratory symptoms and decreased lung functions	LUNN ET AL., 1967

TABLE II.1.-2 QUANTITATIVE COMMUNITY HEALTH EPIDEMIOLOGICAL STUDIES ON EFFECTS OF EXPOSURE TO POLLUTANTS MIXTURES ON PULMONARY FUNCTION

cont'd 3

Pollutants	Measure	Exposure concentrations ug/m^3	Study population	Effect	Reference
		48 - 169 BS 94 - 253 SO_2	Children	No observed effect on respiratory symptoms and lung function	LUNN ET AL., 1970
		170 BS 125 SO_2	Adults	Increased respiratory symptoms	SAWICKI, 1972
		100 - 200 BS 60 - 140 SO_2	Adults	Increased respiratory symptoms	WHO, 1979
O_x and others	lowest estimated effect level	0.10 to 0.15 ppm O_x 0.10 ppm O_x	Young adults	Symptoms of eye irritation, cough, chest discomfort, and headache related to oxidant concentration but not carbon monoxide, nitrogen dioxide, or daily minimum temperature	HAMMER ET AL., 1974
O_x + NO_2	Los Angeles: Median hourly NO_2	130 NO_2 0.07 ppm O_x	205 office workers in Los Angeles	No differences in most tests. Smokers in both cities showed greater changes in pulmonary function than non-smokers.	LINN ET AL., 1976
	90th percentile NO_2	250 NO_2 0.15 ppm O_x			
	San Francisco: Median hourly	65 NO_2 0.08 ppm O_x	439 office workers in San Francisco		
	90th percentile	110 NO_2 0.08 ppm O_x			

TABLE II.2.1.-1 EFFECTS OF MIXTURES OF SO_2 AND O_3 ON PLANTS

Concentration in ug/m^3		Exposure duration	Species	Species effect	Reference
O_3	SO_2				
100	67	6 hr	E. white pine	No effect to needle elongation. Foliar injury on sensitive clones only; 10 trees with 75-100 % of needles with tip necrosis. SO_2 alone caused tip necrosis on 75-100 % of the needles on 1 tree. O_3 alone caused no injury.	HOUSTON, 1974
100	134	8 hr/d. 5 d/wk 18 days	Soybean	Additive foliar injury effects, greater than additive root dry weight	TINGEY ET AL. 1973
100 to 200	270 to 1350	4 hr	Alfalfa Broccoli Cabbage Radish Tomato Tobacco W_3	Greater than additive foliar injury at 0.10 ppm of each gas for alfalfa, broccoli, and radish. Less than additive effect for tomato. At 675 ug/m^3 SO_2 + 200 ug/m^3 O_3 greater than additive injury noted on alfalfa, radish, and tobacco. At 1350 ug/m^3, SO_2 and 100 ug/m^3 O_3 greater than additive injury on broccoli and tobacco and less than additive injury on alfalfa. At 1350 ug/m^3 SO_2 and 200 ug/m^3 O_3 greater than additive effects on alfalfa, cabbage, radish, tobacco.	TINGEY ET AL., 1973
100	340	3 hr	Trembling Aspen (5 clones)	Greater than additive foliage injury to 3 clones, no injury due to SO_2 alone	KARNOSKY, 1976
100	930	3 hr	Trembling Aspen (5 clones)	Greater than additive foliage injury to 4 clones	KARNOSKY, 1976

TABLE II.2.1.-1 EFFECTS OF MIXTURES OF SO_2 AND O_3 ON PLANTS

cont'd 1

Concentration in ug/m^3		Exposure duration	Species	Species effect	Reference
O_3	SO_2				
540	640	2 hr	Tobacco Bel-W3 Tobacco Bel-B Consolidation 402	9-38 % foliar injury - no injury due to either pollutant singly	MENSER and HEGGESTAD, 1966
560	760	4 hr	Tobacco Bel-W3 Tobacco Bel-B Consolidation 402	23-76 % foliar injury - no injury due to either pollutant singly	MENSER and HEGGESTAD, 1966
100	675	4 hr	Alfalfa Onion Soybean Tobacco Bel-B Tobacco White gold	Only tobacco Bel W_3 showed greater than additive foliar injury	TINGEY ET AL., 1973
200	2700		Tobacco Bel-W_3 Lima bean Broccoli Bromegrass Cabbage Radish Spinach Tomato	At 2700 ug/m^3 SO_2 tobacco Bel B and Bel W_3 exhibited greater than additive effects, and there were less than additive effects for Bromegrass, cabbage, spinach, and tomato.	

TABLE II.2.1.-1 EFFECTS OF MIXTURES OF SO_2 AND O_3 ON PLANTS

cont'd 2

Concentration in ug/m^3		Exposure duration	Species	Species effect	Reference
O_3	SO_2				
280	670	6 hr/day 5 days/wk	Scotch Pine	Less than additive foliar effects - no effects due to O_3 alone	NIELSEN ET AL, 1977
580	670	6 hr/day 5 days/wk	Scotch Pine	Less than additive foliar effects - no effect due to O_3 alone	NIELSEN ET AL., 1977
800	1064	4 hr/day 1 time	Apples (Vance Delicious Imperial McIntosh, Golden Delicious)	Foliar injuries, partly increased, partly decreased compared to effects of O_3 alone	SHERTZ ET AL., 1980
800	1064	4 hr/day only 1 time for O_3	Grapes (Ives, Belaware)	Greater than additive foliar injuries	SHERTZ ET AL., 1980
300	400	6 hr/day O_3 4 hr/day SO_2 for 5 days	Radish Cucumber Soybean	Greater than additive foliar injury for radish and cucumber, but no combined effect on soybean	BECKESON and HOFSTRA 1979
260	1064	4 hr 1 time	Pea	Greater than additive foliar injury	OLSZYK and TIBBITTS 1981

TABLE II.2.1.-1 EFFECTS OF MIXTURES OF SO_2 AND O_3 ON PLANTS

cont'd 3

Concentration in ug/m^3		Exposure duration	Species	Species effect	Reference
O_3	SO_2				
100	134	8 hr/d, 5d/wk 5 wks	Radish	Plant weight reductions, additive (leaf fresh and dry weight) or significantly less than additive (plant fresh wt., root fresh and dry weight).	TINGEY ET AL., 1971
100	134	8hr/d, 5d/wk 4 wks	Tobacco	Additive growth reductions	TINGEY and REINERT 1975
		8hr/d, 5d/wk until control plants 40-45 cm high	Alfalfa	less than additive growth reductions	
100	160	7hr/day 68 days	Alfalfa	No significant alteration of plant responses (carbohydrate, protein, dry weight) compared to the effects of single pollutants	NEELY ET AL., 1977
500	670	4hr/d, 3d/wk 1 wks	Soybean	Additive growth effects	REINERT and WEBER, 1980
300 or 900	1200	4 hr	Radish	Additive growth effects	TINGEY and REINERT, 1975
400	530	O_3-4hr/day, 2 days/wk, 8 wks SO_2-4hr/day, 2days/wk, 8 wks	Tomato (Walter)	Greater than additive growth reduction for large single fruits but no additive effect on total fruit	SHEW ET AL., 1982

TABLE II.2.1.-1 EFFECTS OF MIXTURES OF SO_2 AND O_3 ON PLANTS

cont'd 4

Concentration in ug/m^3		Exposure duration	Species	Species effect	Reference
O_3	SO_2				
500	1350	O_3-4hr/day every 6 days for 4 times	Begonia (Schwabenland Red)	No additive effect on flower weight due to SO_2 compared to O_3 alone	REINERT and NELSON, 1980
500	1350	SO_2-4hr/day every 6 days for 4 times	(Wisper '0' Pink)	Less than additive effect on flower weight for Wisper '0' Pink	
500	1350		(Fantasy)	Greater than additive effect on flower weight for Fantasy and Turo	
500	1350		(Renaissance)	No additive effect on Renaissance	
500	1350		(Turo)		
130	400	O_3-11 hr/day avg, 3 mo SO_2-6 hr/day 5 day/wk,5 wk	Snap bean (BBL 290) (BBL 274) (Astro)	Greater than additive effect on green pod weight	HEGGESTAD and BENNETT, 1981
200	270	O_3 and SO_2 6 hr/day, once a week for 12 weeks	Tall fescue (Alta)	Less than additive effect on tillers	FLAGLER and YOUNGER, 1982
400	270			Greater than additive effect on tillers	
600	270				
200	270			Greater than additive effect on top dry weight	
400	270				
600	270				
100	135	O_3-6 hr/day 68 days SO_2-24 hr/day 68 days	Alfalfa (Mesa-Sirsa)	No additive effect on foliage dry weight due to SO_2 compared to O_3 alone.	NEELY ET AL., 1977

TABLE II.2.1.-1 EFFECTS OF MIXTURES OF SO_2 AND O_3 ON PLANTS

cont'd 5

Concentration in ug/m^3		Exposure duration	Species	Species effect	Reference
O_3	SO_2				
300	200 to 1596	5 or 10 days	White bean Soybean	Less than additive growth reductions and foliar injury less than additive foliar injury	HOFSTRA and ORMROD, 1977
200	270	6 hr/d 133 days	Soybean	SO_2 alone and in the mix did not significantly affect the yield and injury responses	HEAGLE ET AL., 1974
500	1340	4hr/day 4 times, 6 days apart	Begonia (5 cultivars)	Less than additive effects for flower weight of one cultivar 1350 ug/m^3 SO_2 alone significantly reduced flower production in the absence of foliar injury for one cultivar	REINERT and NELSON, 1980

TABLE II.2.2.-1 YIELD RESPONSES OF SELECTED TREE SPECIES TO A COMBINATION OF 200 ug/m^3 OZONE AND 190 ug/m^3 (1) NITROGEN DIOXIDE

Species	Response to height and top dry wt % reduction from control (2)(3) (negative unless otherwise noted)		
	O_3	NO_2	O_3 + NO_2
Loblolly Pine	17	15	39
	19	18	16
Loblolly Pine	25	11	24
	9	10	4
Pitch Pine	14	16	26
	+ 14	20	11
Virginia Pine	11	13	23
	2	1	1
Sweetgum	27	32	28
	30	25	19
White Ash	20	+ 5	16
	37	1	37
Green Ash	19	+ 1	22
	17	10	29
Willow Oak	5	10	14
	+ 1	24	13

Source: KRESS and SKELLY, 1982

(1) Exposure Duration 6hr/day over 28 days
Plants were exposed in continously stirred tank reactor exposure chambers in a greenhouse.

(2) Concentrations of the combination were the same as the single gases.

(3) First figure corresponds to height growth, second figure to top dry weight.

TABLE II.2.3.-1: PLANT RESPONSE TO SULFUR DIOXIDE AND NITROGEN DIOXIDE MIXTURES.

Concentration in ug/m^3		Species	Plant Age (weeks)	Exposure duration (hours)	Species % injury	Response Synergism (1)
SO_2	NO_2					
2000	1440	Avena sativa L.	4 - 5	1 or 3	0-5	+
2000	1440	Beta vulgaris var cicla L.	4 - 5	1 or 3	0-5	+
2000	1440	Lathyrus odoratus L.	4 - 5	1 or 3	0-5	+
2000	1440	Raphanus sativus L.	4 - 5	1 or 3	5-8	+
400 - 675	190 - 380	A. sativa	2 - 3	4	0	*
400 - 675	190 - 380	R. sativus	2 - 3	4	0	*
400 - 675	190 - 380	Phaseolus vulgaris L.	2 - 3	4	0	*
1350	850	R. sativus	4 - 5	1 or 3	0-5	+
270	190	Nicotiana tabacum L.	7 - 8	4	0-10	+
1350 - 1890	290 - 400	Orzopsis hymendoides (R&S) Ricker	*	2	16	0
1350 - 1890	290 - 400	Populus tremuloides Michx.	*	2	1	0
1350 - 1890	290 - 400	Sphaeralcea munroana Spach.	*	2	31	0
4050	28800	P. vulgaris	3 - 4	1.17	70-75	+
6210	24960	Lycopersicon esculentum Mill.	3 - 5	1	35-85	+
6210	23040	Cucumis sativus L.	3 - 4	0.67	50-100	+
6480	24960	A. sativa	3 - 4	1	40-75	-
6480	28800	Capsicum frutescens L.	5 - 6	1	10-58	+
135 - 675	96 - 480	P. vulgaris Pinto	3 - 4	4	0-24	+
135 - 675	96 - 480	A. sativa	3 - 4	4	0-27	+
135 - 675	96 - 480	R. sativus	3 - 4	4	0-27	+
135 - 675	96 - 480	Glycine max (L.) Merr.	3 - 4	4	0-35	+
135 - 675	96 - 480	N. tabacum	7 - 8	4	0-18	+
135 - 675	96 - 480	L. esculentum	5 - 6	4	0-17	+

(1) +, Greater than additive; 0, additive; -, less than additive, * Not defined

TABLE II.2.3.-2: NON-ADVERSE EFFECT LEVELS OF SENSITIVE PLANT SPECIES UNDER SENSITIVE CONDITIONS FOR SO_2 + NO_2 DURING SEVERAL EXPOSURE TIMES.

		SO_2 in ug/m^3				
		< 40	100	250	500	1000
	< 40	> 100 D	30 D	15 H	3,5 H	1,4 H
	100	> 100 D	10 H	4,0 H	2,0 H	1,2 H
NO_2	250	7 D	5 H	3,1 H	1,8 H	1,2 H
in	500	38 H	3,8 H	2,5 H	1,7 H	1,2 H
ug/m^3	1000	13 H	3 H	2,1 H	1,7 H	1,2 H

D = Days H = Hours

TABLE II.2.5.-1: EFFECT ON YIELD OF VARIOUS PLANT SPECIES DUE TO EXPOSURE TO OZONE, SULPHUR DIOXIDE, AND NITROGEN DIOXIDE

Concentration in ug/m^3			Species	Exposure duration	Response	Yield, % reduction from control (negative unless otherwise noted)							Reference
O_3	SO_2	NO_2				O_3	SO_2	NO_2	SO_2+NO_2	O_3+SO_2	O_3+NO_2	$O_3+SO_2+NO_2$	
380	400	290	Snap Bean	4hr, 3 times/wk 4 wks	Green bean fresh wt	27	9	+12	20	6	25	27	REINERT and HECK, 1982
600	800	580	Marigold	3hr/day, 3 days/wk, 3 wks	Flower wt	20	47	+16	13	23	+ 4	20	REINERT and SANDERS, 1982
600	800	580	Marigold	3hr/day, 3 days/wk, 1wk	Flower wt	41	49	23	47	25	39	20	SANDERS and REINERT, 1982
600	800	580	Radish	3hr/day, 3 days/wk, 1wk	Hypocotyl	30	+21	+10	+16	43	33	65	SANDERS and REINERT, 1982
800	1080	770	Radish	3hr + 6hr 1 time	Hypocotyl	20	4	0	13	24	23	36	REINERT and GRAY, 1981
500	675	480	Azalea	3hr/day, 6 times in a 4-wk period	Foliage	6	7	0	17	22	16	27	SANDERS and REINERT, 1982

TABLE II.2.5.-1: EFFECT ON YIELD OF VARIOUS PLANT SPECIES DUE TO EXPOSURE TO OZONE, SULPHUR DIOXIDE, AND NITROGEN DIOXIDE

Cont'd 1

Concentration in ug/m^3			Species	Exposure duration	Response	Yield, % reduction from control (negative unless otherwise noted)				Reference
O_3	SO_2	NO_2				O_3	SO_2	NO_2	$O_3+SO_2+NO_2$	
300	400	290	Kentucky bluegrass (12 cultivars)	O_3-hr/day, 10 days SO_2-cont. 10 days NO_2-, continuous, 10 days	Leaf area	5	12	6	16	ELKIEY, 1980
			Red top grass	O_3-hr/day, 10 days SO_2-cont. 10 days NO_2-continuous, 10 days		14	12	12	28	
			Creeping bentgrass	O_3-hr/day, 10 days SO_2-cont. 10 days NO_2-continuous, 10 days		7	18	8	26	
			Colonial bentgrass	O_3-hr/day, 10 days SO_2-cont. 10 days NO_2-continuous, 10 days		15	6	13	27	

TABLE II.2.5.-1: EFFECT ON YIELD OF VARIOUS PLANT SPECIES DUE TO EXPOSURE TO OZONE, SULPHUR DIOXIDE, AND NITROGEN DIOXIDE

cont'd 2

Concentration in ug/m^3			Species	Exposure duration	Response	Yield, % reduction from control (negative unless otherwise noted)				Reference
O_3	SO_2	NO_2				O_3	SO_2	NO_2	$O_3+SO_2+NO_2$	
			Red fescue (2 cultivars)	O_3-hr/day, 10 days SO_2-cont. 10 days NO_2-continuous, 10 days		16	0	0	22	
			Perennial Ryegrass	O_3-hr/day, 10 days SO_2-cont. 10 days NO_2-, continuous, 10 days		20	+7	2	13	

TABLE II.3-1 AIR POLLUTION DAMAGE TO MATERIALS (1)

Materials	Type of damage	Principal air pollutants	Other environmental factors
Metals	Corrosion, tarnishing	Sulfur oxides and other acid gases	Moisture, air, salt, micro-organisms
Building stone	Surface erosion and discoloration	Sulfur oxides and other acid gases, particulate matter	Moisture, temperature fluctuations, salt, vibration, micro-organisms, CO_2
Paint	Surface erosion, discoloration	Sulfur oxides, hydrogen sulfide, ozone, particulate matter	Moisture, sunlight, microorganisms
Textiles	Reduced tensile strength, soiling	Sulfur oxides, nitrogen oxides, particulate matter	Moisture, sunlight, physical wear
Textile dyes	Fading, colour change	Nitrogen oxides, ozone	Sunlight
Paper	Embrittlement	Sulfur oxides	Moisture, physical wear
Rubber	Cracking	Ozone	Sunlight, physical wear
Leather	Weakening, powdered surface	Sulfur oxides	Physical wear
Ceramics	Changed surface appearance	Acid gases, HF	Moisture, micro-organisms

(1) Adapted from YOCOM and UPHAM, 1977.

Table III.2.2.-1: Mean concentrations of sulphur dioxide and nitrogen oxides, as measured by the EMEP-network in the sampling period April 1982 - September 1982, in ug/m^3

Country	Station code and name	SO_2	NO_2	Height above sea level (m)
Switzerland	CH1 Jungfraujoch	1.8		3573
Czechoslovakia	CS2 Chopok	4.8		2008
German Federal Republic	D3 Schauinsland	3.0	1.8	1205
	D5 Brotjacklriegel	7.0	1.8	1016
Denmark	DK7 Færøerne-Akraberg	0.4		90
France	F3 La Crouzille	2.4		460
	F6 Valduc	2.0		470
Hungary	H1 K-puszta	1.6	2.0	125
Norway	N1 Birkenes	0.8	0.8	190
	N36 Hummelfjell	0.2		1539
	N37 Bjørnøya	0.2		20
Portugal	P1 Braganca	3.0		691
Sweden	S5 Bredkälen	2.0	0.3	404
Finland	SF4 Ähtäri	2.6		162
United Kingdom	UK2 Eskdalemuir	4.4		243

Table III.2.3.-1: SO_2-concentration in ug/m^3 as measured in 1981 at urban sites within the EC

Country	min. value	max. value	50 % value	98 % value
Belgium	4	414	71	242
	2	171	32	118
	2	185	20	109
	2	274	25	150
	4	218	36	181
	10	688	68	237
	20	243	76	195
	10	184	47	147
	3	249	61	167
	8	377	78	185
	8	445	76	247
	3	147	30	92
	2	373	31	196
	3	295	40	177
	10	271	58	175
	2	218	35	130
	5	234	48	152
	4	233	56	165
	8	266	53	120
	4	278	49	180
	8	233	64	161
	8	315	62	199
	8	210	64	158
	15	427	81	227
	4	290	54	161
	4	203	41	148
	2	288	39	181
	2	235	34	114
	5	188	39	140

Table III.2.3.-1 cont'd 1

Country	min. value	max. value	50 % value	98 % value
Federal Republic of Germany	10	470	70	380
	10	380	60	280
	10	780	80	360
	10	480	70	320
	10	670	60	290
	10	500	60	240
	13	130	14	90
	13	212	17	84
	13	231	25	113
	12	421	64	305
	10	736	61	299
	13	252	35	200
	13	208	35	157
	13	224	29	182
	13	157	31	123
	13	234	15	84
	13	243	38	199
	10	270	40	160
	8	633	51	241
	10	540	50	210
	10	390	40	220
	7	539	41	273
	13	220	30	162
	13	253	28	140
	13	303	45	190
	13	261	31	164
	13	108	31	97
	13	256	22	187

Table III.2.3.-1 cont'd 2

Country	min. value	max. value	50 % value	98 % value
Denmark	3	103	18	64
	1	104	7	84
	3	152	18	98
France	7	341	53	209
	13	479	75	279
	5	352	53	225
	4	439	65	295
	8	368	51	196
	5	250	46	156
	1	302	34	167
	1	207	29	151
	4	271	51	169
	5	116	28	90
	4	96	31	81
	8	104	31	90
	3	127	37	96
	1	218	19	162
	1	228	13	108
	1	394	11	276
	1	527	25	320
	2	741	65	432
	1	599	32	417
	1	391	22	154
	1	336	46	200
	1	212	33	169
	1	428	55	248
	1	420	20	233
Ireland	10	320	88	209
	5	211	44	109
	6	110	50	86

Table III.2.3.-1 cont'd 3

Country	min. value	max. value	50 % value	98 % value
Italy	10	1141	104	941
Netherlands	1	313	23	109
	2	124	22	94
	1	320	38	141
	3	298	26	106
	2	314	18	128
	1	322	11	71
	1	329	12	107
	4	191	21	114
	1	318	23	108
	3	240	23	135
	2	228	21	106
	1	377	19	104
United Kingdom	2	369	29	116
	6	232	39	104
	7	76	22	51
	6	72	22	57
	14	284	84	202
	8	89	29	60
	9	257	41	128
	6	278	41	135
	12	455	55	195
	20	313	74	216
	13	267	60	182
	20	234	64	169
	7	275	36	190
	7	326	62	211
	13	229	40	166
	12	355	57	224
	15	387	51	180

Table III.2.3.-1 cont'd 4

Country	min. value	max. value	50 % value	98 % value
United Kingdom cont'd	11	204	33	111
	12	296	48	191
	6	369	34	159
	7	210	54	159
	6	174	36	132
	7	189	46	137
	6	232	34	123
	6	101	18	72
	6	117	29	100
	5	68	18	48
	8	94	27	70
	13	226	52	125
	7	137	36	105
	17	116	36	94
	6	229	29	84

Table III.2.3.-2: Results of SO_2-measurements in ug/m^3 carried out within the GEMS-network of the WHO in 1980

Country	City	arith. mean	98 %	max. value
Belgium	Brussels 1	76	200	327
	2	69	199	359
	3	64	186	272
Denmark	Copenhagen 1	21	84	318
	2	16	78	109
	3	26	105	141
France	Paris 1	95	270	466
	2	87	273	382
	3	81	252	330
F.R. Germany	Frankfurt 1	74	196	245
Greece	Athens	44	198	340
Ireland	Dublin 1	56	133	202
	2	34	81	157
	3	23	57	113
Italy	Milan 1	242	939	1279
	2	167	449	728
Netherland	Amsterdam 1	26	66	123
	2	30	98	215
	3	28	80	165
Portugal	Lisbon 1	44	105	170
	2	65	195	270

Table III.2.3.-2 cont'd 1

Country	City	arith. mean	98 %	max. value
Spain	Madrid 1	63	227	303
	2	30	135	163
	3	86	388	651

Table III.2.3.-3: SO_2-values in ug/m^3 measured in the period 1982 to 1984 in German cities

Station	measurement period (April-March)	annual values 50% value	annual values 98% value	Winter values 50 % value	
Berlin-Marienfelde	1982/83	49	159	1982/83	61
	1983/84	44	198	1983/84	70
Berlin-Jungfernheide	1982/83	67	200	1982/83	99
	1983/84	61	268	1983/84	90
Duisburg	1982/83	49	234	1982/83	56
	1983/84	58	239	1983/84	78
Dortmund	1982/83	48	187	1982/83	57
	1983/84	53	198	1983/84	66
Köln	1982/83	37	167	1982/83	45
	1983/84	34	136	1983/84	42
Witten	1982/83	36	160	1982/83	41
	1983/84	42	126	1983/84	54
Essen	1982/83	37	170	1982/83	40
	1983/84	42	179	1983/84	52
Gelsenkirchen	1982/83	62	242	1982/83	71
	1983/84	62	230	1983/84	82
München	1982/83	27	84	1982/83	34
	1983/84	26	72	1983/84	29
Mannheim	1982/83	38	130	1982/83	38
	1983/84	30	150	1983/84	40
Hannover	1982/83	23	157	1982/83	23
	1983/84	25	225	1983/84	41
Frankfurt	1982/83	34	133	1982/83	44
	1983/84	49	166	1983/84	66

Table III.2.4-1: SO_2-concentrations in ug/m^3 at rural sites in three Member States of the EC, as measured in 1982

Country	min. value	max. value	50 % value	98 % value
F.R. Germany	1	169	9	113
	1	133	10	90
	1	151	8	84
	1	82	4	42
	1	104	3	28
	1	137	4	57
	1	352	12	118
	1	156	14	81
	1	139	12	93
	1	154	4	57
	1	150	3	27
	1	208	11	107
	1	250	12	135
The Netherlands	3	276	20	110
	1	285	19	126
	1	319	13	103
	1	271	7	98
United Kingdom	4	56	11	23
	7	153	34	115
	7	102	23	52
	6	44	13	22
	4	210	22	108
	12	162	40	116

Table III.2.6.1.-1: Trends in SO_2-concentrations at urban site for 1976 to 1981

Station code	SO_2-concentrations in ug/m^3						
	1976	1977	1978	1979	1980	1981	1982
02101 016	75	114	110	110	100	80	n.a.
03201 334	40	42	30	27	30	23	24
06101 009	n.a.	78	73	n.a.	109	78	78
08303 418	46	41	31	45	44	38	n.a.
01201 001	139	142	107	94	77	71	60
01301 809	117	105	100	104	87	73	72
01401 509	93	116	96	84	71	51	43
01402 709	135	109	77	101	98	64	60
01403 215	55	71	55	60	57	75	62
04302 015	61	95	41	53	50	34	n.a.
04402 032	80	95	45	55	58	73	n.a.
09102 111	128	129	108	105	89	75	n.a.
09103 413	60	70	59	65	53	38	n.a.
09201 020	77	79	70	74	74	58	n.a.
09302 048	80	68	70	52	58	42	n.a.

n.a.: not available

Table III.2.6.2.-1 SO_2-trend analysis for five rural sites in the F.R. Germany for the period 1973 to 1982

Station code	annual mean	summer mean	winter mean	percentiles 20	30	60	70	95	98
WES	++	++	=	=	+	+	=	-	--
WAL	+	=	=+	=+	+	+	+	++	+
DEU	+	=	=+	=	=	=	=+	+	++
SCH	-	--	=	-	-	-	-	-	=
BRO	=	=	=	-	-	=-	=-	=	=

= : no changes in concentrations (correlation coefficient $r < 0.2$)

=+/=-: slightly increasing/decreasing concentrations ($r = 0.2 \ldots 0.4$)

+/- : increasing/decreasing concentrations ($r = 0.4 \ldots 0.7$)

++/--: clear increase/decrease in concentrations ($r > 0.7$)

Table III.2.8.-1: Black Smoke concentrations in ug/m^3 as measured in 1981 in some Member States of the European Communities

Country	min. value	max. value	50 % value	98 % value
Belgium	3	126	22	77
	2	62	12	42
	1	74	8	32
	1	82	8	53
	1	91	14	69
	1	81	13	52
	8	108	33	85
	1	83	11	49
	1	81	14	48
	1	147	14	66
	1	83	10	50
	3	69	14	48
	1	194	14	54
	1	132	19	77
	3	81	10	42
	3	81	12	48
	3	98	14	51
	2	106	14	70
	2	51	10	34
	2	143	22	102
	2	62	14	45
	2	74	12	54
	2	97	12	45
	1	118	19	68
	1	71	12	38
	1	74	10	48
	1	78	18	67
	1	122	16	69
	1	155	23	88

Table III.2.8.-1: cont'd 1

Country	min.value	max. value	50 % value	98 % value
Denmark	2	103	22	62
	1	130	9	42
	1	111	10	54
	2	146	19	78
France	5	392	32	161
	6	296	33	135
	6	287	34	134
	6	281	31	115
	10	385	39	143
	2	104	22	83
	1	121	23	83
	2	87	17	68
	2	116	19	80
	1	104	9	54
	1	74	10	51
	1	40	5	28
	1	30	4	23
	2	104	23	59
	1	97	17	65
Ireland	1	706	24	212
	1	576	19	171
	2	445	21	198
	1	124	13	81
United Kingdom	3	149	21	77
	2	121	16	81
	4	183	19	73
	3	136	20	70
	1	125	13	67
	1	116	9	56
	3	287	23	141

Table III.2.8.-1: cont'd 2

Country	min. value	max. value	50 % value	98 % value
United Kingdom	2	246	18	109
cont'd 1	3	541	18	148
	4	650	13	142
	3	323	10	159
	3	372	26	119
	1	174	7	50
	1	134	13	70
	1	107	8	53
	2	88	14	62
	3	140	16	101
	1	105	7	55
	1	310	22	129
	3	287	22	154
	4	230	29	164
	3	164	15	85
	1	111	8	55
	1	129	14	78
	3	227	18	87
	4	285	21	108
	2	71	8	36
	1	99	12	45
	3	122	26	103
	8	492	38	238
	2	87	9	59
	1	94	14	69
	1	193	10	69

Table III.2.8.-2: Black Smoke concentration in ug/m^3 as measured in 1980 by the WHO within the GEMS network

Country	City		arith. mean	98 %	max. value
Belgium	Brussels	1	19	51	92
		2	17	53	90
		3	15	47	77
Denmark	Copenhagen	1	24	57	66
		2	14	23	57
		3	16	49	78
France	Paris	1	44	134	194
		2	51	137	199
	Toulouse	1	26	99	203
		2	18	65	180
Greece	Athens (1)	1	202	369	464
		2	218	342	369
Ireland	Dublin	1	41	166	269
		2	14	50	99
		3	16	59	169
United Kingdom	Glasgow	1	36	172	365
		2	21	131	253
		3	33	224	328
	London	1	20	69	102
		2	25	95	205
		3	84	171	296
Spain	Madrid	1	130	312	395
		2	56	220	310

(1) data incomplete

Table III.2.8.-3: Black Smoke concentration in ug/m^3 as measured at a number of urban sites in the F. R. Germany

Station	measurement period	annual values		winter values
		50 % value	98 % value	50 % value
Berlin-Marienfelde	1982/83	13	64	19
	1983/84	13	91	26
Berlin-Jungfernheide	1982/83	14	71	21
	1983/84	15	106	32
Berlin-Schöneberg	1982/83	24	158	42
	1983/84	28	152	51
Duisburg	1982/83	13	54	15
	1983/84	15	93	20
Dortmund	1982/83	17	63	19
	1983/84	17	91	22
Köln	1982/83	15	45	15
	1983/84	15	73	21
Witten	1982/83	14	53	15
	1983/84	17	80	22
Essen	1982/83	8	34	8
	1983/84	12	61	13
Gelsenkirchen	1982/83	15	80	19
	1983/84	19	117	27
München	1982/83	39	123	40
	1983/84	37	137	37
Mannheim	1982/83	15	63	19
	1983/84	17	94	26
Karlsruhe	1982/83	12	53	19
	1983/84	14	85	26
Hannover	1982/83	8	54	12
	1983/84	9	98	18
Frankfurt	1982/83	15	57	19
	1983/84	19	98	28

Table III.2.9.-1: Black Smoke concentration in ug/m^3 at rural sites, as measured 1981 in the United Kingdom

Country	min. value	max. value	50 % value	98 % value
United Kingdom	1	61	1	20
	1	126	9	64
	1	38	2	25
	1	35	3	17
	1	27	2	14
	1	57	6	36
	1	95	4	35
	1	4	1	3

Table III.2.10.-1: Trends in Black Smoke concentrations at a number of urban sites within the EC.

Station code	Black Smoke concentrations in ug/m^3						
	1976	1977	1978	1979	1980	1981	1982
01201 001	24	18	18	19	19	20	21
01301 809	55	51	44	43	35	36	27
01401 509	26	14	17	14	16	12	7
01402 709	24	14	8	8	14	14	12
01403 215	16	20	17	18	18	14	n.a.
03201 334	7	7	9	7	13	12	13
04101 017	n.a.	n.a.	33	31	30	33	n.a.
04302 015	42	55	25	22	24	23	n.a.
09102 111	24	26	26	24	24	19	n.a.
09103 413	31	27	24	26	22	23	n.a.
09201 020	28	33	29	23	23	19	n.a.
09302 048	37	34	29	25	33	27	n.a.

n.a.: not available

Table III.2.11.-1: Concentration ranges of SO_2 and Black Smoke as measured in urban and industrial areas within the Member States of the European Community (1)

Member State	Area/City	24 h-mean value in ug/m^3		Annual mean in ug/m^3	
		SO_2	BS	SO_2	BS
Belgium	Brussels, Anvers, Liège, Charleroi, Gand	80...300	30...70	20...90	10...50
Denmark	Kobenhavn	40...120	5...40	5...35	5...25
F. R. Germany	Berlin, Ruhr-area, Rhein-area, Hamburg, Ludwigshafen, Frankenthal, Mainz, Saarbrücken, Neunkirchen, Billingen, Völklingen, Kassel, Wetzlar, Untermain-area, Rhein-Main-area, Stuttgart, Nürnberg-Erlangen, München, Ingolheim, Würzburg	200...400 =========	20...120 ===	70...120	10...40
France	Paris, Strasbourg, Gravenchou, Lettarve, Bouches-de-Rhône, Lille, Vitry-sur-Seine	80...260	30...120	30...90	15...80
Greece	Athens, Thessaloniki	80...120	estim.20...130	50...60	estim.10...40
Ireland	Dublin, Cork, Dundalk, Limerick, Drogheda	60...240 ===	20...240 ===	20...90	10...50
Italy	Milano, Torino, areas of Piemonte, Ligerrio, Toscana, Lusio, Compania, Sicilia	150...300 ===	estim.20...150 ===	50...160	estim.10...40
Luxembourg	Colmar-Burg	50...100	25...50	20...35	10...20
The Netherlands	Rijnmond area	50...175	35...100	5...35	5...25
United Kingdom	London areas in Yorkshire, Northern Wales, North-West West-Midlands, East-Midlands, Scotland	100...240 =========	80...350 ===	50...100 ========	30...80 =======

(1) Values with a low probability of exceeding the non-adverse effect levels are underlined once, such with a high probability of exceeding the non-adverse effect level are underlined twice.

TABLE III.3.3.-1: NO_2-concentrations in ug/m^3 within some Member States of the EC at urban sites

Country	City	Year 19'	1 hour values		24 hours values	
			50 %	98 %	50 %	98 %
Belgium	Zellik	79-80	57	133	63	103
	Zellik	80-81	63	219	63	158
	Bruxelles	79-80	63	219	63	201
	Bruxelles	80-81	63	156	63	133
	Antwerpen	79-80	67	211	70	171
	Antwerpen	80-81	51	99	50	80
F. R. Germany	Frankfurt	80	57	154	54	129
	Frankfurt	81	55	129	63	103
	Köln	80	49	122	51	106
	Köln	81	44	150	53	141
France	Paris	81	76	152	72	118
	Paris	80-82	67	162	68	125
Netherlands	Haarlem	81-82	50	118	48	104
	Nijmegen	81-82	53	114	51	104
	Badhoevedorp	81-82	48	108	44	49
	Delft	80	38	101	38	89
	Hoevelaken	81-82	38	93	38	86
United Kingdom	Islington	77	50	108	51	87
	Islington	78	40	118	42	93
	Harrow	79-80	38	106	40	91
	Stevenage	80	32	82	34	69
	Stevenage	81	34	101	34	82
	Canvey	78	38	95	34	74
	Canvey	79	27	78	30	61
	London CLL	80	61	125	63	110
	London CLL	81	67	135	67	112

Table III.3.4.-1: NO_2-concentrations in ug/m^3 at rural sites

Country	Site	Year	1 Hour Values		24 Hours Values	
			50 %	98 %	50 %	98 %
F. R. Germany	Deuselbach	1980	9	36		
		1981	9	42		
	Langenbruegge	1980	8	30		
		1981	10	40		
Netherlands	Brandwijk	81-82	34	84	32	80
	Hem	81-82	15	76	19	68
	Biddinghuizen	81-82	21	74	23	72
	Sellingen	81-82	15	63	15	61
United Kingdom	Sibton	1978	11	49	13	38
	Sibton	1979	8	36	8	27

Table III.3.6.2.-1: Results of the UBA-trend analysis for 5 rural sites

	annual mean	summer mean	winter mean	Percentiles 20	30	60	70	95	98
WES	=+	=	=	=	=	=+	+	=	=
WAL	++	++	=+	+	+	+	+		=+
DEU	+	=	+	=	=	=+	=+	++	++
SCH	++	++	=+	+	+	++	++	+	+
BRO	=+	=	=+	=	=	=	=	=+	=+

= : no changes in concentrations (correlation coefficient $r < 0.2$)

=+/=-: slightly increasing/decreasing concentrations ($r = 0.2 \ldots 0.4$)

+/- : increasing/decreasing concentrations ($r = 0.4 \ldots 0.7$)

++/--: clear increase/decrease in concentrations ($r > 0.7$)

Table III.3.7.-1: Concentration ranges of SO_2 and NO_2 at different site-categories as measured within the Member States of the European Community

	24 h-mean value in ug/m^3		annual mean-value in ug/m^3	
	urban	rural	urban	rural
SO_2	40 ... 350	10 ... 150	40 ... 160	5 ... 30
NO_2	40 ... 300	20 ... 60	20 ... 100	5 ... 30

Table III.3.7.-2: Relative number of days at which selected threshold-values for NO_2 and SO_2 have been exceeded at rural sites in the Netherlands and the F.R. Germany

Site	Threshold concentration in ug/m^3		days of exceedance in %		
	NO_2	SO_2	summer	winter	total
Nieuw-Namen (1)	>30	>40	9.6	31.3	20.5
	>40	>40	6.0	26.4	16.2
Den Helder (1)	>30	>40	1.8	9.7	5.8
	>40	>40	1.2	8.3	4.8
Biddinghuizen (1)	>30	>40	1.5	11.3	6.4
	>40	>40	0.8	9.2	5.0
Puiflijk (1)	>30	>40	3.7	22.1	12.9
	>40	>40	2.5	20.0	11.3
Kloosterburen (1)	>30	>40	0.8	11.0	5.9
	>40	>40	0.5	7.4	3.9
Deuselbach (2)	>15	>15			18.0
	>30	>30			2.0
	>20	>40			9.0
Schauinsland (2)	>15	>15			0.2
	>30	>30			n.d.

(1) hourly values from 01.04.1984 to 31.03.1985

(2) 1/2 h values for SO_2 and O_3, daily for NO_2 from 01.01.1984 to 31.12.1984

n.d. not detected

III.4.2.-1: Background concentrations of O_3 at ground levels

Location	Period	concentrations and averaging times	Reference
Greenland	July 1960	25 ug/m^3 instantaneous maximum	MCKEE (1961)
Antarctica	April 1957 to May 1958	20 to 67 ug/m^3 mean monthly avg. at surface	ODISHAW (1959)
Geographic South Pole	1961 to 1964	40 to 80 ug/m^3 monthly mean	ALDAZ (1967)
	1963 and 1964	20 to 60 ug/m^3 monthly mean	

III.4.2.-2: Background concentrations of O_3 at mountain sites

Location	Period	Altitude	O_3 in ug/m^3		References
			Range	Average	
Arosa, Switzerland	1950-59	1860 m	19-90	50	COETZ AND VOLZ (1951)
Mt. Capilio and	1951-52	3100 m	18-85	45	REGENER (1957)
Albuquerque, New Mexico		1600 m	3-120	36	
Mauna Loa Observatory, Hawaii	1954	3000 m	30-62	45	RICE AND PALES (1959)
Zugspitze, Germany	1977-82	3000 m	30-140	80	FRICKE (1983)

Table III.4.3.-1: O_3-concentration in ug/m^3 measured at urban sites

Country	Station	Period	Annual average	98 % (95 %)	max. value
F. R. Germany	Frankfurt	1976-1982	28		410
	Köln-Godorf	1976-1981	24		546
	Bonn	1976-1978	24		424
	Freiburg	1981-1982	50		306
	Mannheim-Mitte	1975-1980	28	(112)	
Netherlands	Den Helder	1980/81	65	130 (113)	
	Sellingen		52	133(111)	
	Oost-Maarland		32	117 (92)	
	Nieuw-Namen		49	140 (115)	
France	Northern urban sites	1980-1984	30...40	60...110	
	Southern urban sites		35...60	50...150	
United Kingdom	average over-all sites		20...30		400...500

Table III.4.4.-1: O_3-concentrations in ug/m^3 measured at rural sites in the F. R. Germany

Station	Year 19'	annual arithm. mean	annual 98 %	summer 98 %	max. value
Westerland	80	62	116	124	150
	81	64	122	134	168
	82	58 (*)	98	104	126
Deuselbach	80	62	160	184	290
	81	58	130	148	194
	82	56	148	162	220
	83	54	128	138	184
Waldhof	79	68 (*)	176	192	254
	80	60	158	188	244
	81	38 (*)	146	220	270
	82	54 (*)	160	180	230
	83	84	204	226	320
Schauinsland	80	112	202	218	358
	81	80 (*)	140	138	180
	82	90	170	184	266
	83	90	170	188	154
Brotjacklriegel	80	64	114	122	162
	81	80	150	156	194
	82	84	154	174	208
	83	84	166	174	224

(*) data sets incomplete

Table 4.5.3.-1: Maximum hourly ozone concentration in ug/m^3 during 30th May - 5th June 1982 episode

Country	Station	May		June				
		30	31	1	2	3	4	5
S	Falsterbro	-	160	-	-	186	166	170
	Rörvik	158	140	156	194	218	250	248
N	Jeloya	132	130	108	128	162	190	174
	Langesund	132	130	128	146	184	180	152
DK	OTC	120	144	156	162	194	168	236
UK	Stevenage	192	214	316	168	168	328	272
	Sibton	100	134	112*	168*	170	168	188
NL	Den Helder	-	-	222	206	-	250	210
	Sappemeer	118	160	226	224	230	224	202
	Biddinghuizen	158	196	324	214	282	268	234
	Waarde	180	230	186	276	244	226	310
	Posterholt	156	176	218	230	236	232	254
B	Antwerpen R822	194	220	170	-	290	244	280
	Antwerpen R801	198	190	-	-	162	-	-
D	Deuselbach	116	140	186	184	136	148	152
	Schauinsland	176	182	260	210*	208	234	168
	Brotjacklriegel	134*	196	180*	170*	176	138	178
A	Illmitz	236	230	236	222	202	186	198
	Bildein	148	174	186	180	172	174	170

* Night-time value (23 h - 5 h)

Table III.4.6.1.-1: O_3-concentration in ug/m^3 at three urban sites in the F. R. Germany

Station	Year	annual mean	summer mean
Köln-Eifelwall	1976	32	120
	1977	20	68
	1978	18	60
	1979	22	78
	1980	20	72
	1981	24	78
Karlsruhe-West	1976	38	176
	1977	23	101
	1978	27	80
	1979	26	93
	1980	29	93
	1981	36	109
Mannheim-Nord	1976	32	142
	1977	32	100
	1978	48	165
	1979	27	95
	1980	24	89
	1981	28	110

Table III.4.7.-1: Concentration ranges for SO_2, NO_2 and O_3 in ug/m^3 at different site-categories

Pollutant	Urban sites		Rural sites	
	annual mean	24 h-mean value	annual mean	24 h-mean value
SO_2	40 ... 160	40 ... 350	5 ... 30	10 ... 150
NO_2	20 ... 100	40 ... 300	5 ... 30	20 ... 60
O_3	20 ... 40	60 ... 300	50 ... 80	60 ... 300

Table III.4.7.-2: Relative number of days at which the threshold values for NO_2, O_3 and SO_2 were exceeded at rural sites in the Netherlands and the F. R. Germany

Site	threshold-concentration in ug/m^3			days of exceedance in %		
	NO_2	SO_2	O_3	Summer	Winter	Total
Nieuw-Namen (1)	>30	>50	>30	3	2	<3
Den Helder (1)	>30	>50	>30	1	n.d.	<1
Biddinghuizen (1)	>30	>50	>30	1	n.d.	<1
Puiflijk (1)	>30	>50	>30	1	2	<2
Kloosterburen (1)	>30	>50	>30	n.d.	n.d.	n.d.
Deuselbach (2)	>20 >40	>50 >60	>50 >60			<1 n.d.

(1) hourly values for the period 01.04.84 to 31.03.85

(2) 1/2 h values for SO_2 and O_3, daily values for NO_2 for the period 01.01.84 to 31.12.84

n.d. = not detected

Table IV.3.-1.: Structure of total man-made SO_2-emissions
in kilotonnes and percentages of totals, as estimated for 1983

Source category / Country:	B	DK	F	FRG	GR	IRL	I	L	NL	UK	Total
Power plants	405(53)	210(48)	1240(53)	2010(62)	310(65)	98(45)	2250(62)	2(10)	193(58)	2900(68)	9618(61)
Transport	34(5)	12(3)	150(6)	102(3)	18(4)	5(2)	195(5)	1(4)	12(4)	50(1)	578(36)
Industry	196(26)	157(36)	670(29)	770(24)	126(27)	60(27)	705(19)	17(81)	95(29)	810(19)	3606(23)
Households & others	125(16)	58(13)	290(12)	372(11)	21(4)	56(26)	515(14)	2(10)	29(9)	533(12)	2001(13)
Total	760(5)	437(3)	2350(15)	3254(21)	475(3)	219(1)	3665(23)	21(0,1)	329(2)	4293(27)	15803(100)

Table IV.3.-2.:Structure of total man-made NO_x-emissions
in kilotonnes and percentages of totals, as estimated for 1983

Source category	Country: B	DK	F	FRG	GR	IRL	I	L	NL	UK	Total
Power plants	85(20)	82(32)	240(13)	859(28)	65(43)	28(38)	750(40)	11(10)	79(15)	763(41)	2962(29)
Transport	241(58)	115(45)	1163(63)	1693(55)	57(38)	23(31)	710(37)	52(47)	282(55)	725(39)	5061(50)
Industry	62(15)	31(12)	305(16)	434(14)	21(14)	13(18)	200(11)	39(35)	122(24)	307(17)	1534(15)
Households & others	29(7)	28(11)	145(8)	114(3)	7(5)	10(13)	220(12)	9(8)	33(6)	51(3)	646(6)
Total	417(4)	256(3)	1853(18)	3100(30)	150(1)	74(1)	1880(19)	111(1)	516(5)	1846(18)	10203(100)

Table IV.3.-3: Structure of Total Man-made HC- and Particulate Emissions of EC-Member States in Kilotonnes and Percentage of Totals, as Estimated for 1983

Source Category	Pollutants	
	HC	Particulates
Power generation	250 (3)	790 (22)
Transport	3200 (39)	180 (5)
Refineries	200 (3)	-
Industries	500 (6)	2100 (58)
Domestic & others	1100 (13)	530 (15)
Use of volatile organics	3000 (36)	-
Total	8250 (100)	3600 (100)

Table IV.4.2.-1.: Incremental costs of SO_2-emissions reduction strategies

Control strategies	Costs (in US$ per ton SO_2)	
Coal cleaning	30-150	
Utility strategies[a]		
Fuel Switching		
Shift from high- to low-sulfur coal	250-350	
Shift from high- to medium-sulfur coal	350-400	
Shift from medium- to low-sulfur coal	400-500	
Shift from high- to low-sulfur residual oil	300-400	
Flue gas desulfurization		
Shift from unscrubbed to scrubbed high-sulfur coal	400-600	
Shift from unscrubbed to scrubbed medium-sulfur coal	600-1500	
Shift from unscrubbed to scrubbed low-sulfur coal	1800-3000	
Limestone injection multistaged burners[b]		
High-sulfur coal	250-500	200-350
Medium-sulfur coal	300-1100	250-700
Low-sulfur coal	600-2000	500-1200
Industrial strategies[c]		
Fuel Switching		
Shift from high- to low-sulfur coal	250-350	
Shift from high- to medium-sulfur coal	350-400	
Shift from medium- to low-sulfur coal	400-500	
Shift from high- to low-sulfur residual oil	300-400	
Flue gas desulfurization		
Shift from unscrubbed to scrubbed high-sulfur coal	400-600	
Shift from unscrubbed to scrubbed medium-sulfur coal	600-1500	
Shift from unscrubbed to scrubbed low-sulfur coal	1800-3000	

[a] Representative costs for a 500-MW power plant. Costs will vary for each region and year.

b Removal of SO_2 for retrofits expected to be between 50% (first column) and 60% (second column)

[c] Representative costs for a 170-million Btu/hour industrial boiler. Costs will vary for each region and year

FIGURES

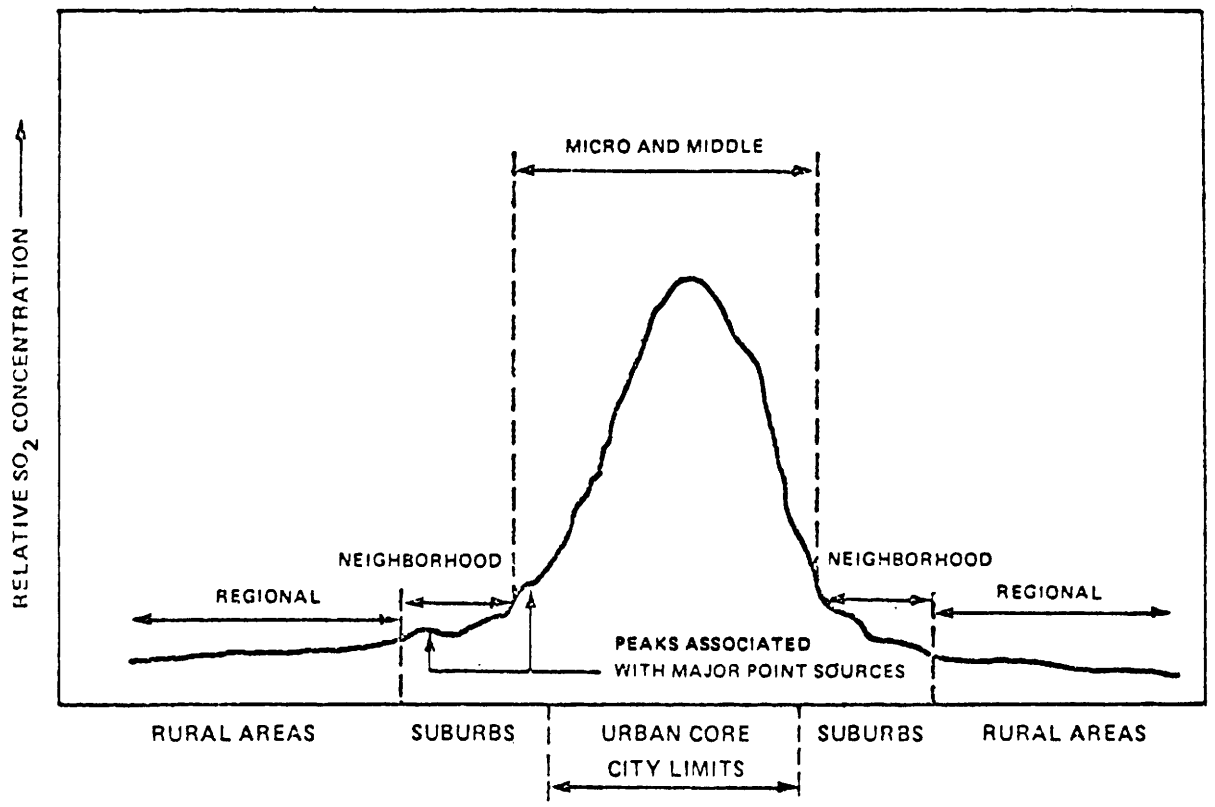


Figure III.1.-1: Relative locations for sites measuring concentrations represent several spatial scales of measurement in an urban complex, with respect to annual averaging times.

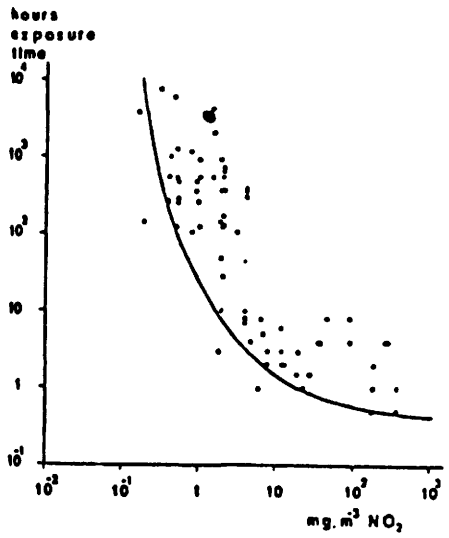


Diagram showing the 2-dimensional concentration/time model for defined effects of NO_2 on plants.
●: effects; ○: no effects

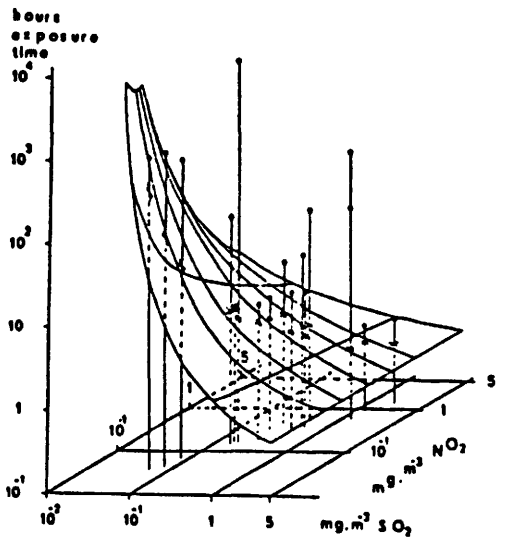
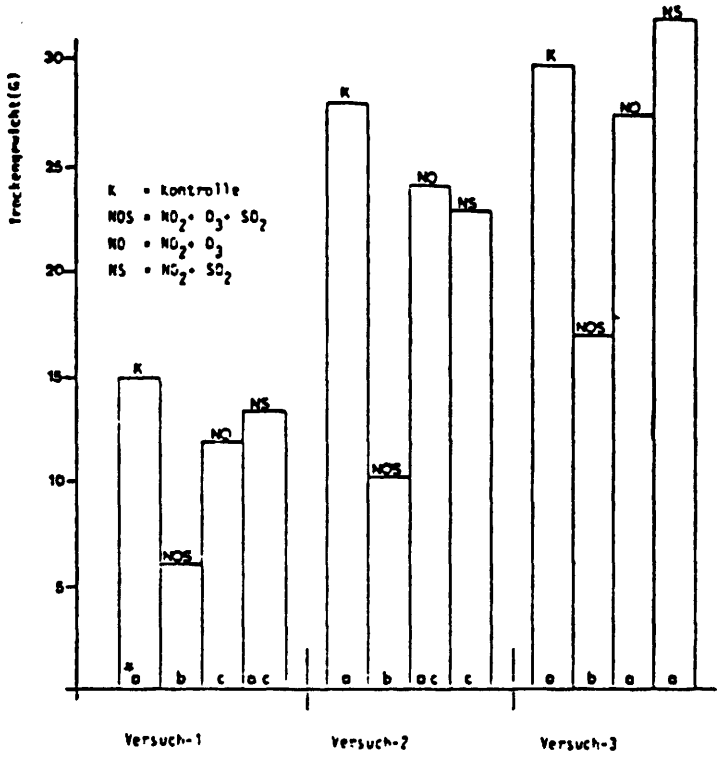


Diagram showing the 3-dimensional concentration/time model for defined effects of NO_2 + SO_2 on plants.
●: effects

Figure II.2.3.-1: Concentration/time models for NO_2 and NO_2 + SO_2



	Versuch 1 16. 2.-30. 3. 1981			Versuch 2 6. 4.-18. 5. 1981			Versuch 3 25. 5.-6. 7. 1981		
µg/m³	NO_2	O_3	SO_2	NO_2	O_3	SO_2	NO_2	O_3	SO_2
Tag	57	55	60	58	62	58	44	54	60
Nacht	23	3	60	32	0	58	31	0	60
Spitzen				93			101		
				9-10 u. 15-16 Uhr			9-10.30 u. 15-16 Uhr		

Bei gleichen Buchstaben besteht kein signifikanter Unterschied zwischen den Gruppen ($P \leq 0.05$)

Abb. 8. Ertrag (T. S.) von *Plantago major*, der mit Mischungen aus SO_2, O_3 und NO_2 begast wurde

Figure II.2.4.-1: Effects of SO_2, NO_2 and O_3 on Plantage major

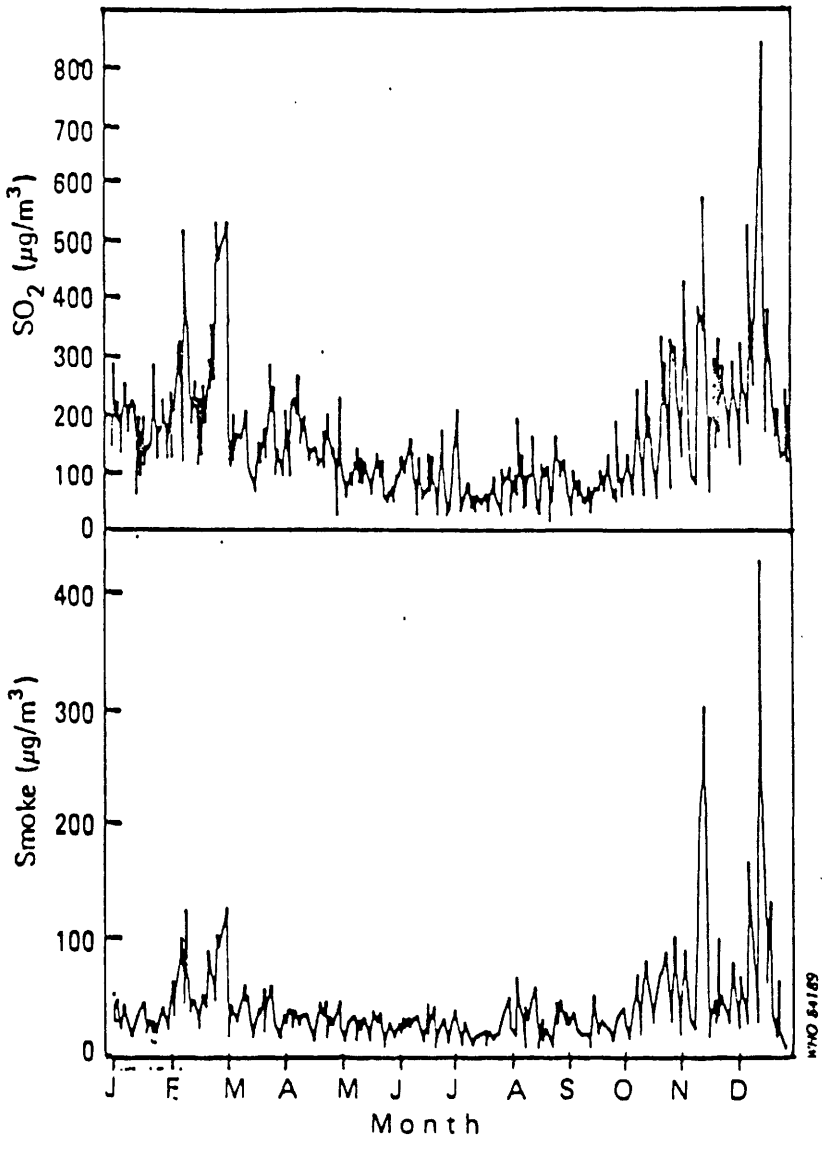


Figure III.2.5.2.-1: Daily records of SO_2 and smoke concentrations in London during 1975 at an urban site

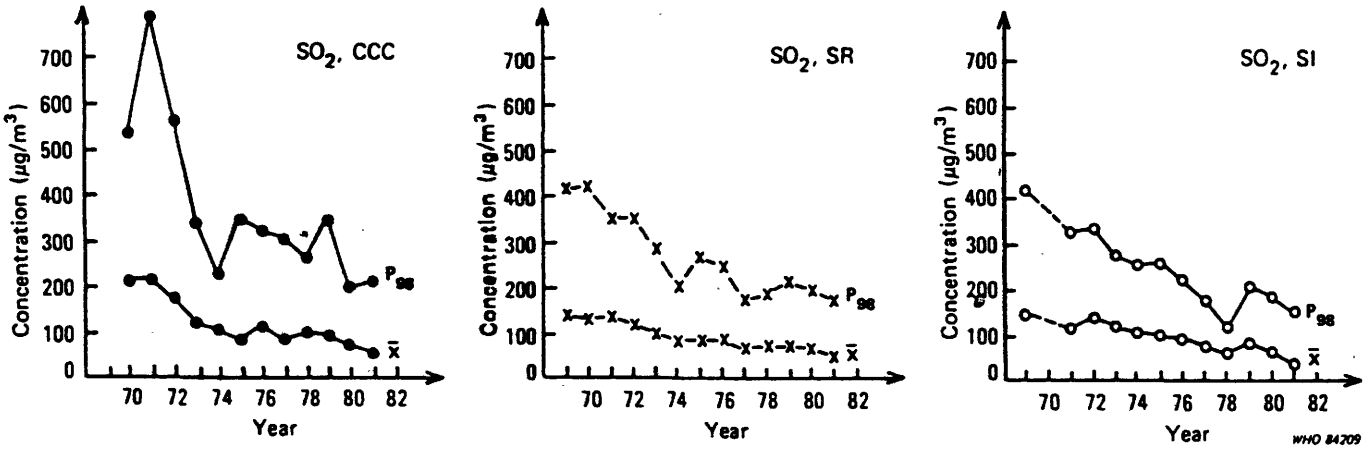


Figure III.2.6.1.-1: Trends in SO_2-concentrations in Brussels for 1969 - 1981 at three urban sites.

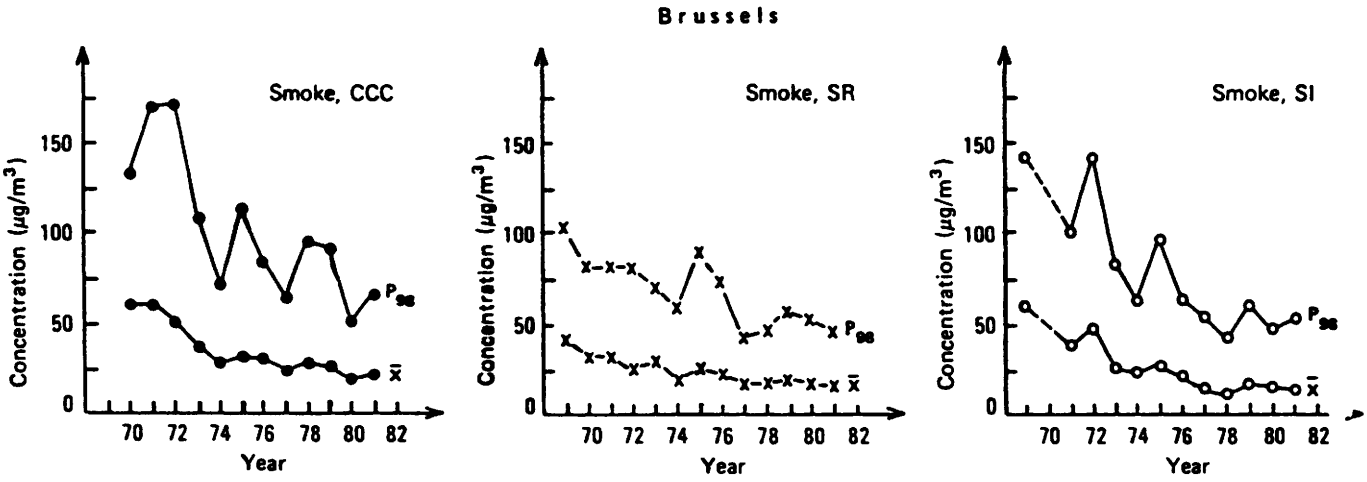


Figure III.2.10.-1: Trends in Black Smoke concentrations in Brussels for 1969 - 1981 at three urban sites.

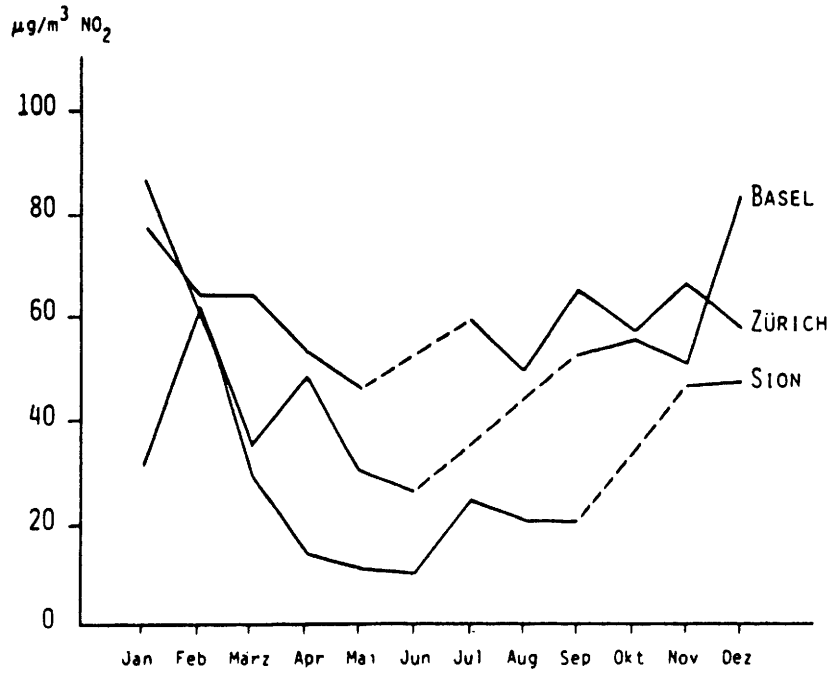


Figure III.3.5.-1: Typical seasonal variations in NO_2-concentration in middle and north Europe

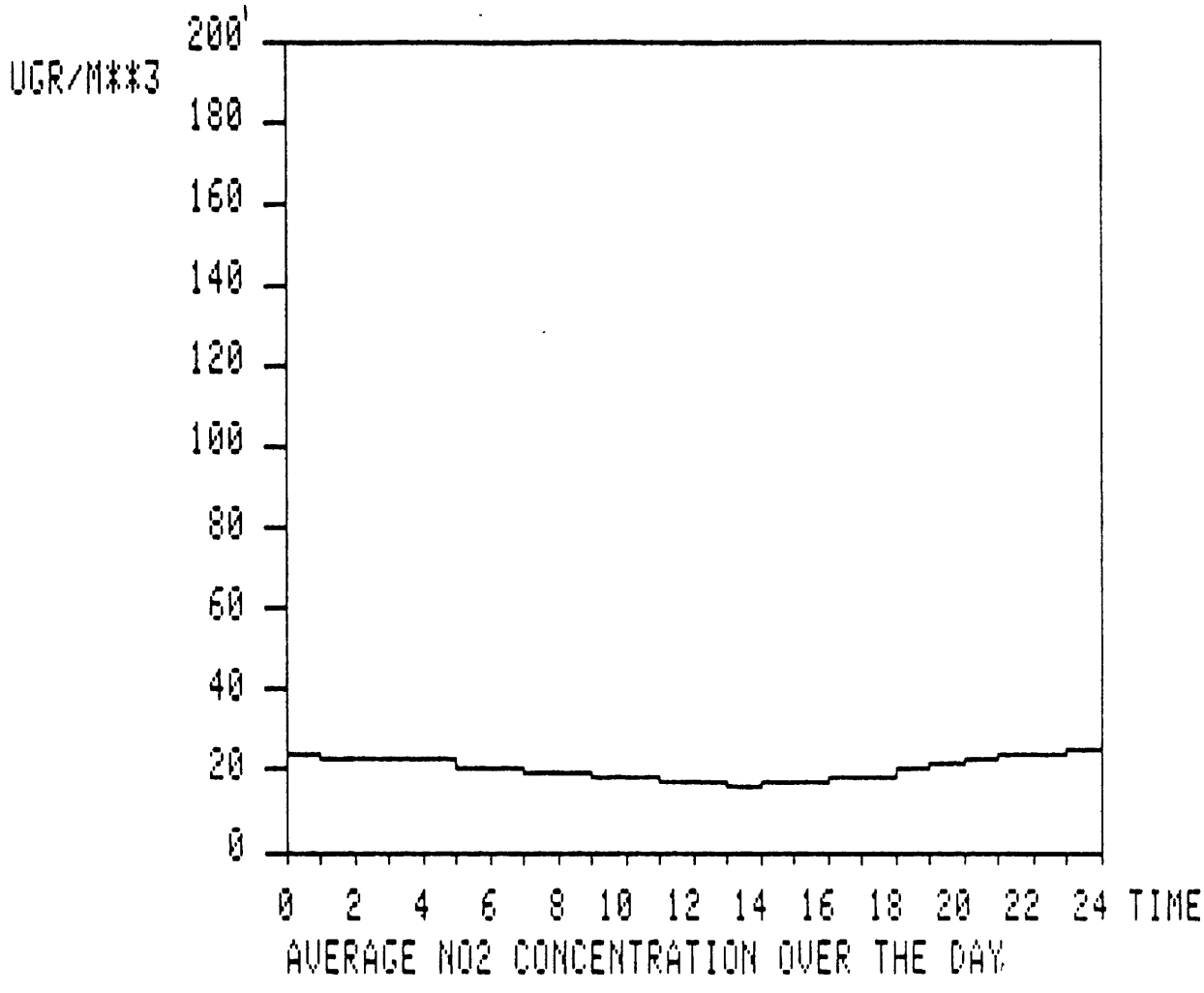


Figure III.3.5.1.-1: Example for a diurnal NO_2-pattern, measured at a rural site in the Netherlands

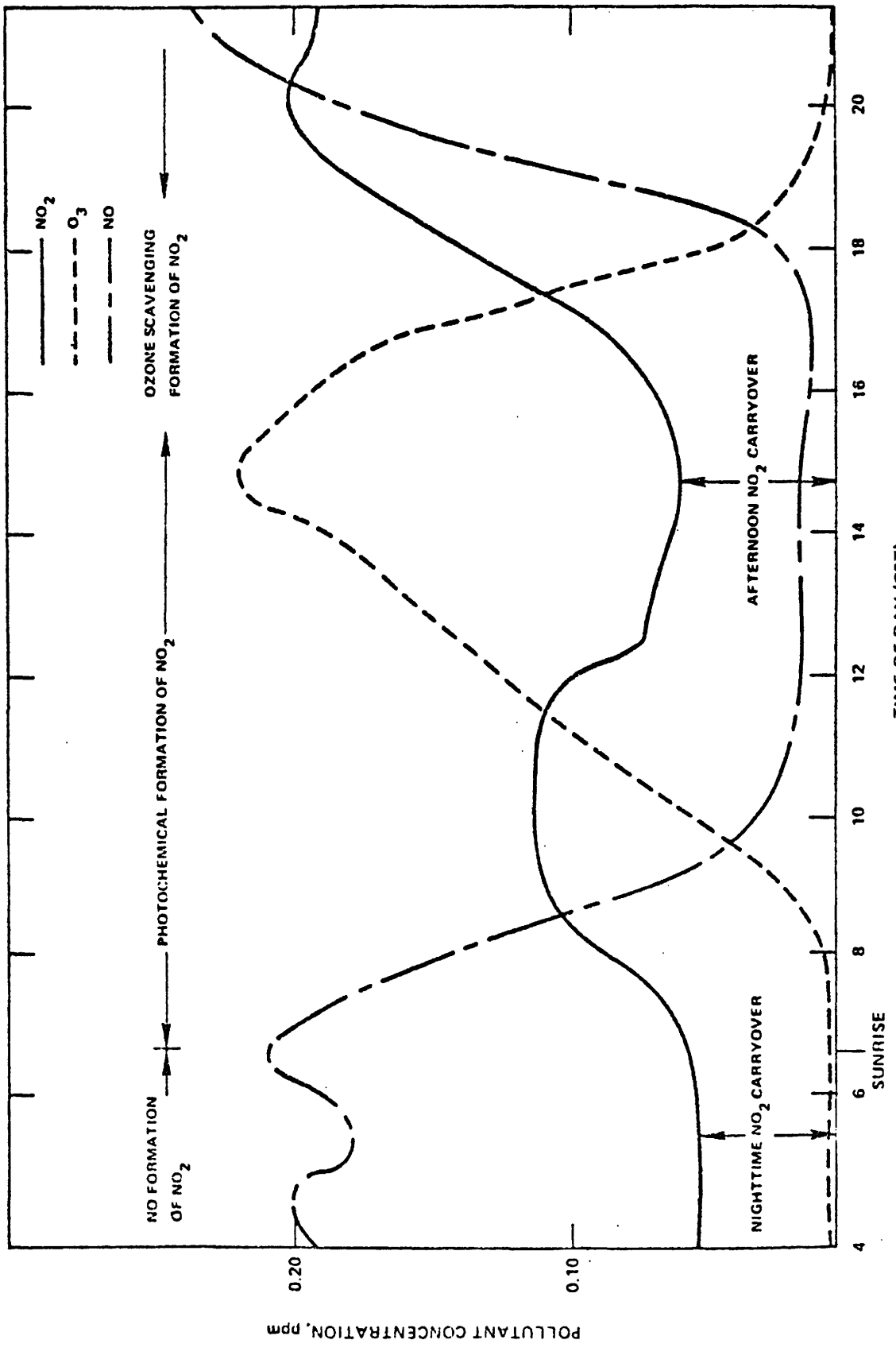


Figure III.3.5.1.-2: NO_2-concentration pattern. Illustration of photochemical and ozone scavenging formation of NO_2.

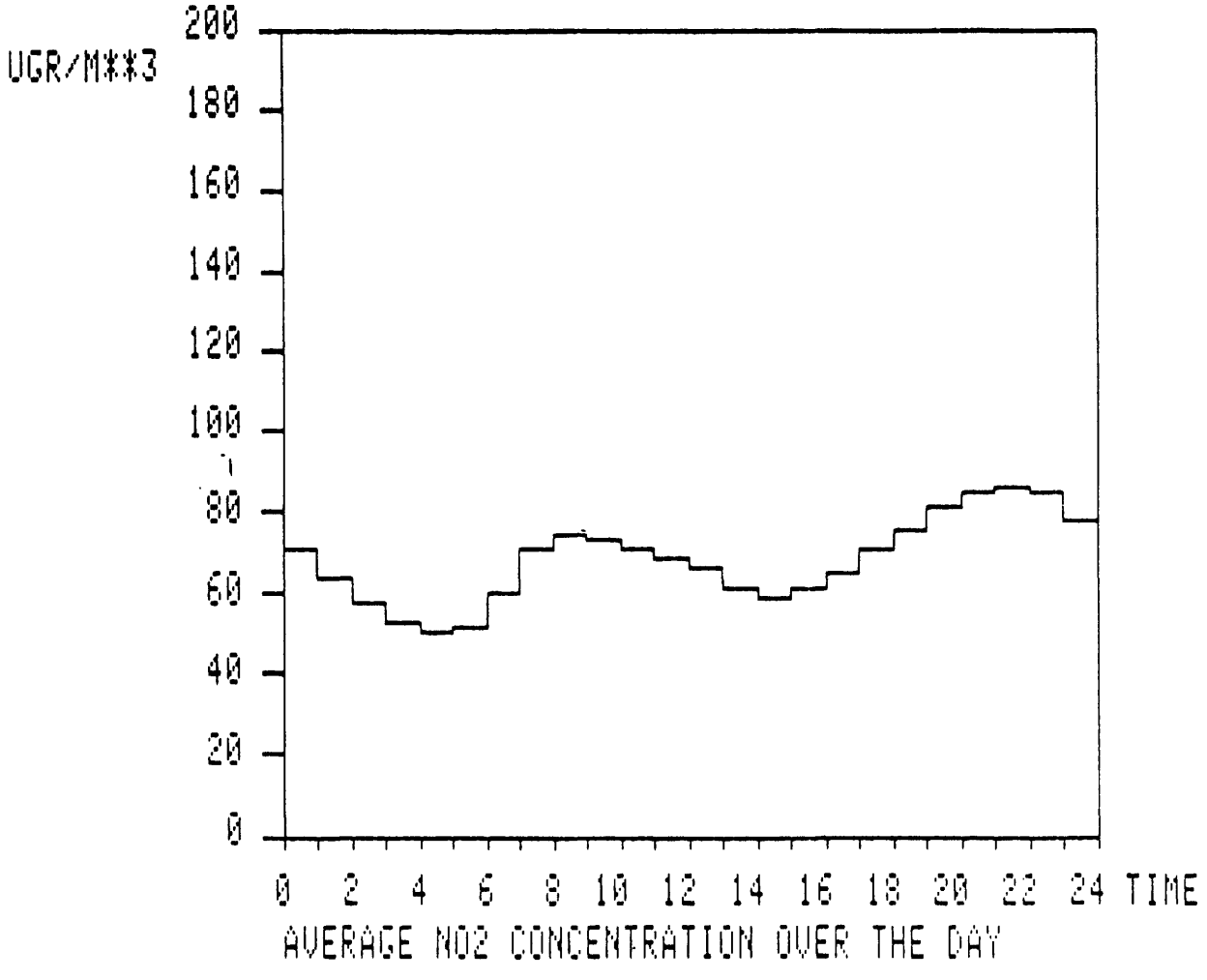


Figure III.3.5.1.-2: Typical pattern for a build-up of NO_2-concentrations caused by traffic emissions in a city without occurance of substantial photochemical reactions.

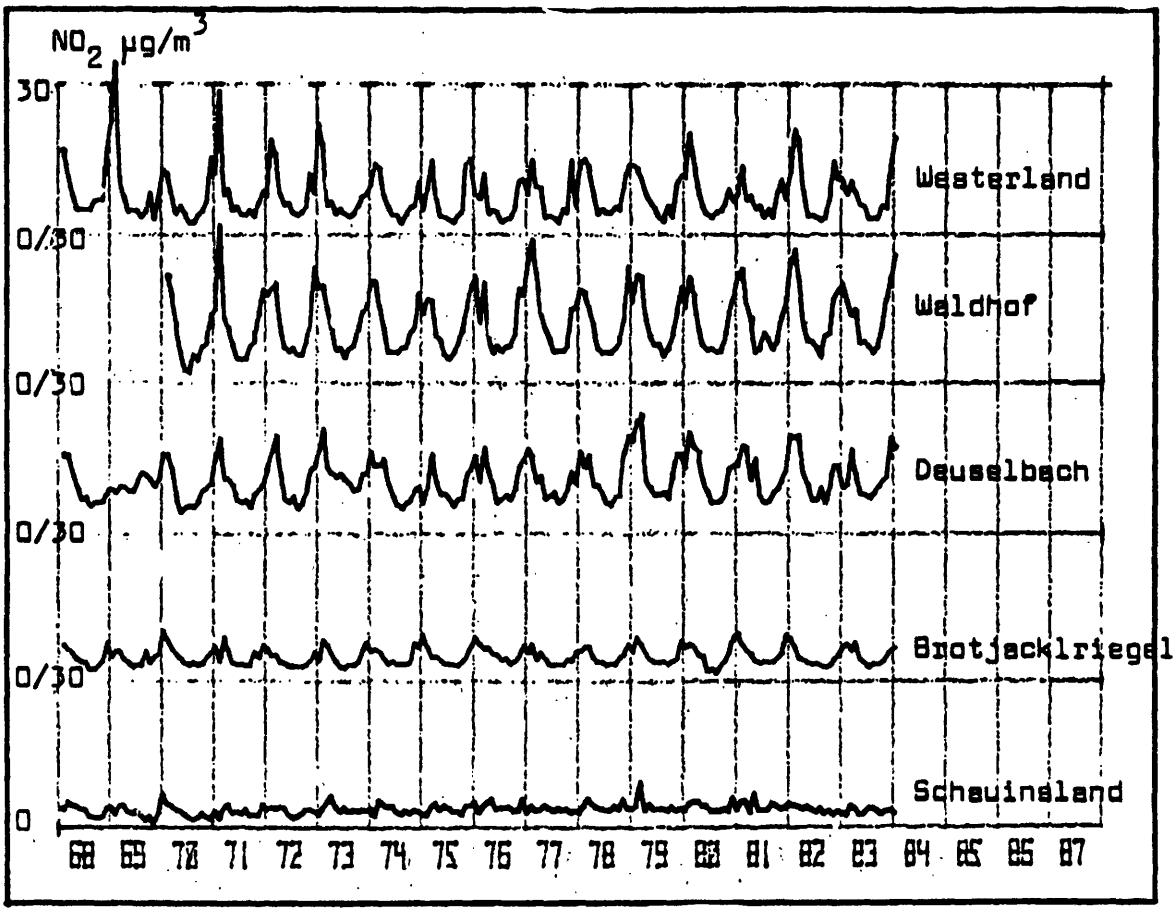


Figure III.3.5.2.-1: Seasonal Variations of NO_2 at rural sites

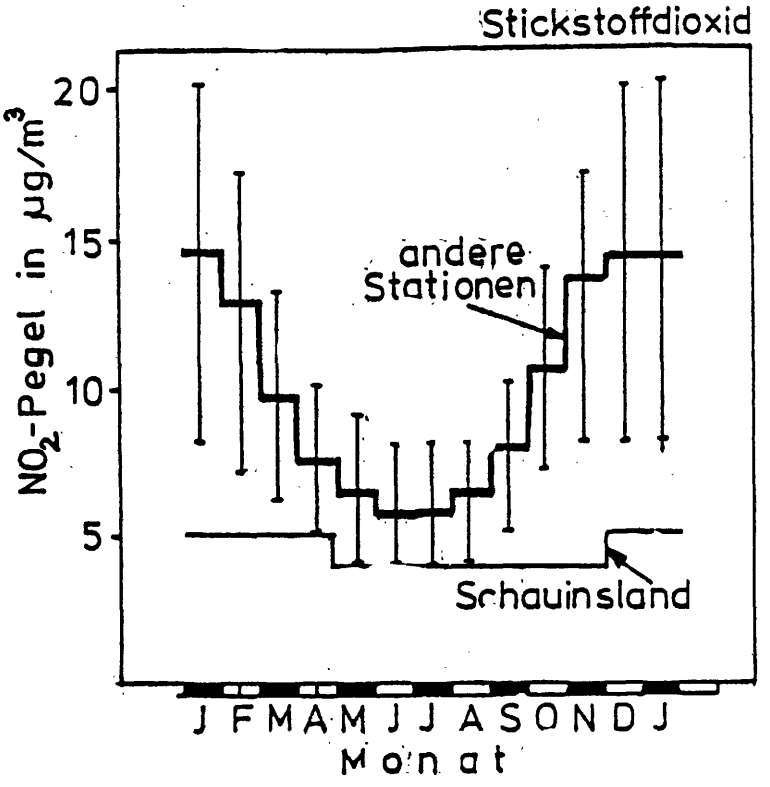


Figure III.3.5.2.-2.: Seasonal patterns of NO_2 at rural sites

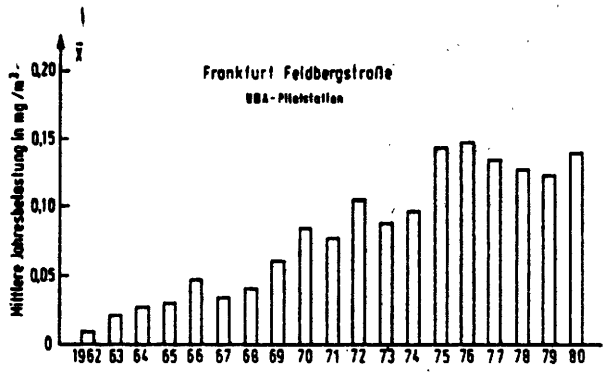


Figure III.3.6.1.-1: NO_2-trend data, measured at Frankfurt (F.R.G.)

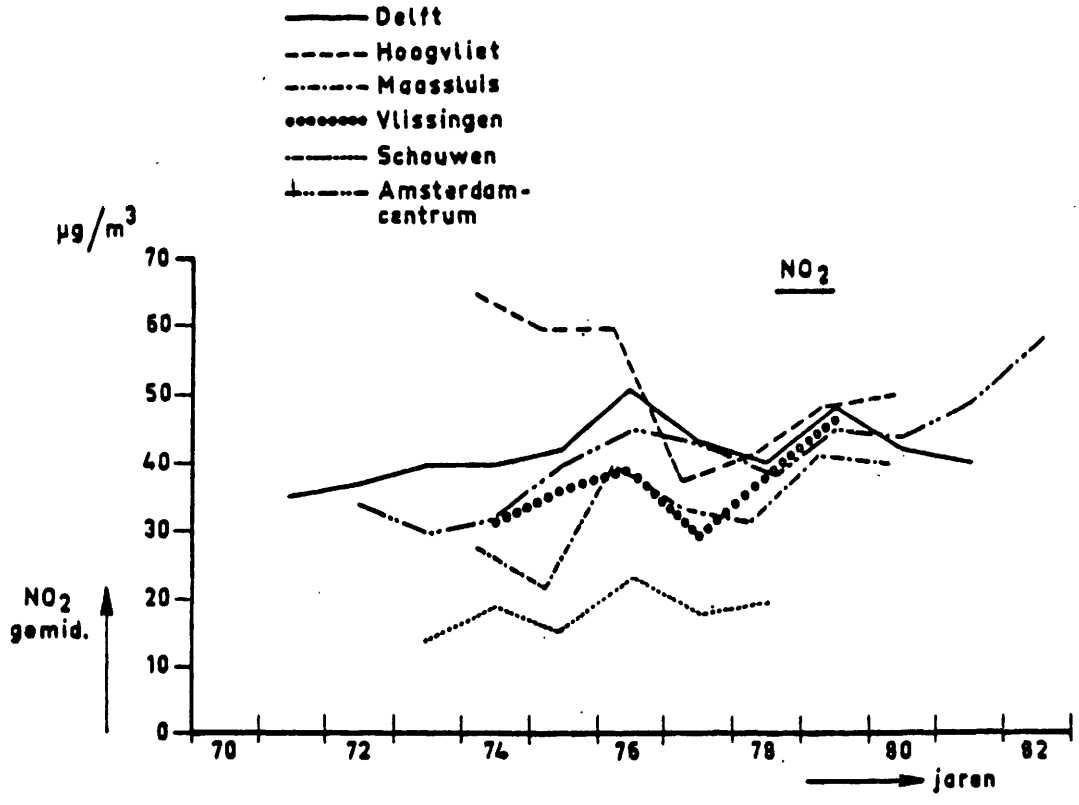


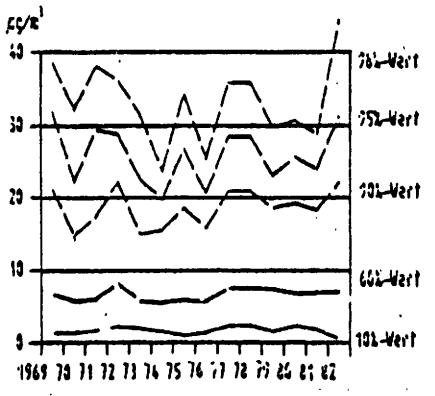
Figure III.3.6.1.-2: NO_2-trend data as measured in several Dutch cities

- Prozentwerte der relativen Summenhaeufigkeits-

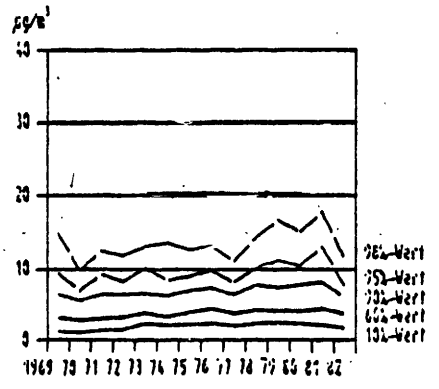
verteilung der Tagesmittelwerte -

Ergebnisse aus dem UBA-Messnetz

in wenig belasteten Gebieten



MESSSTELLE WESTERLAND



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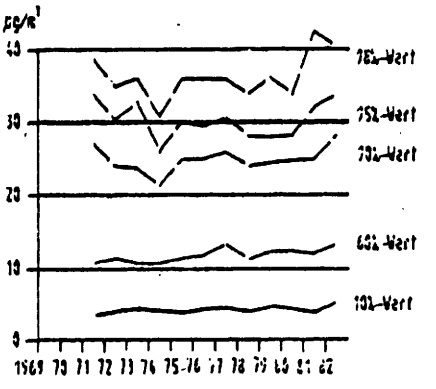

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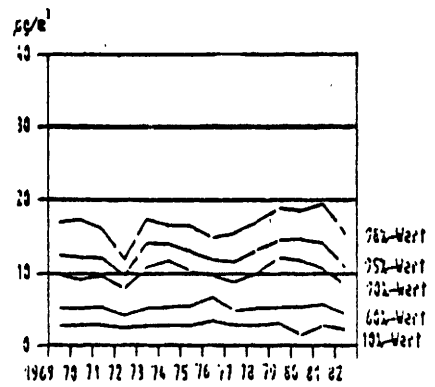

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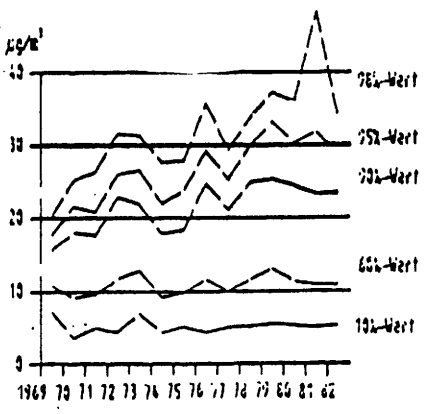

MESSSTELLE DEUSELBACH

Figure III.3.6.2.-1: Trends in NO_2-concentrations at five rural sites in F. R. Germany.

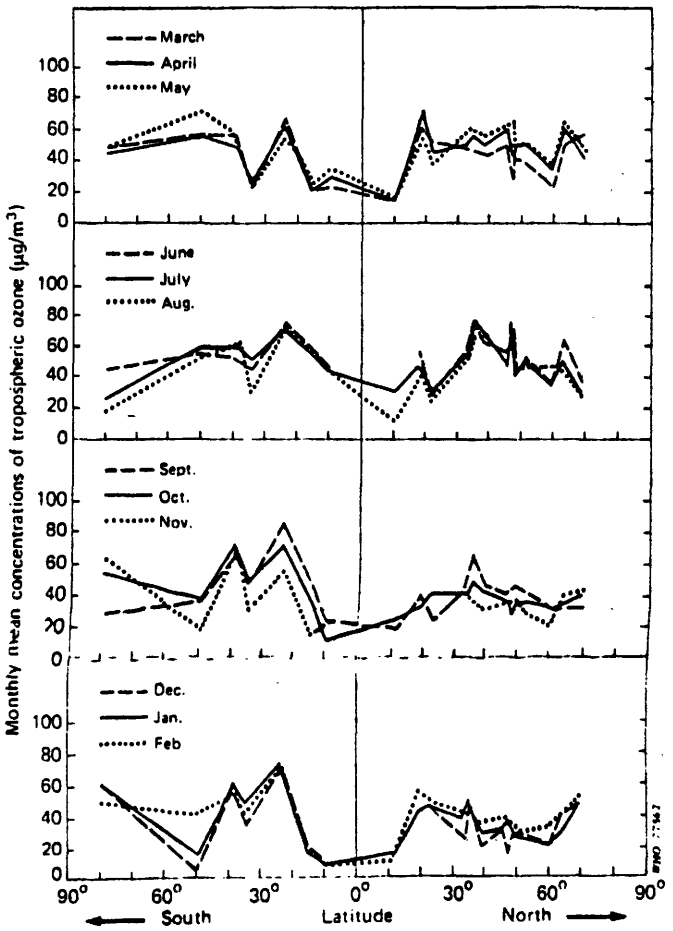

Figure III.4.2.-1: Global distribution of background ozone concentrations (FABIAN & PRUCHNIEWICZ, 1973)

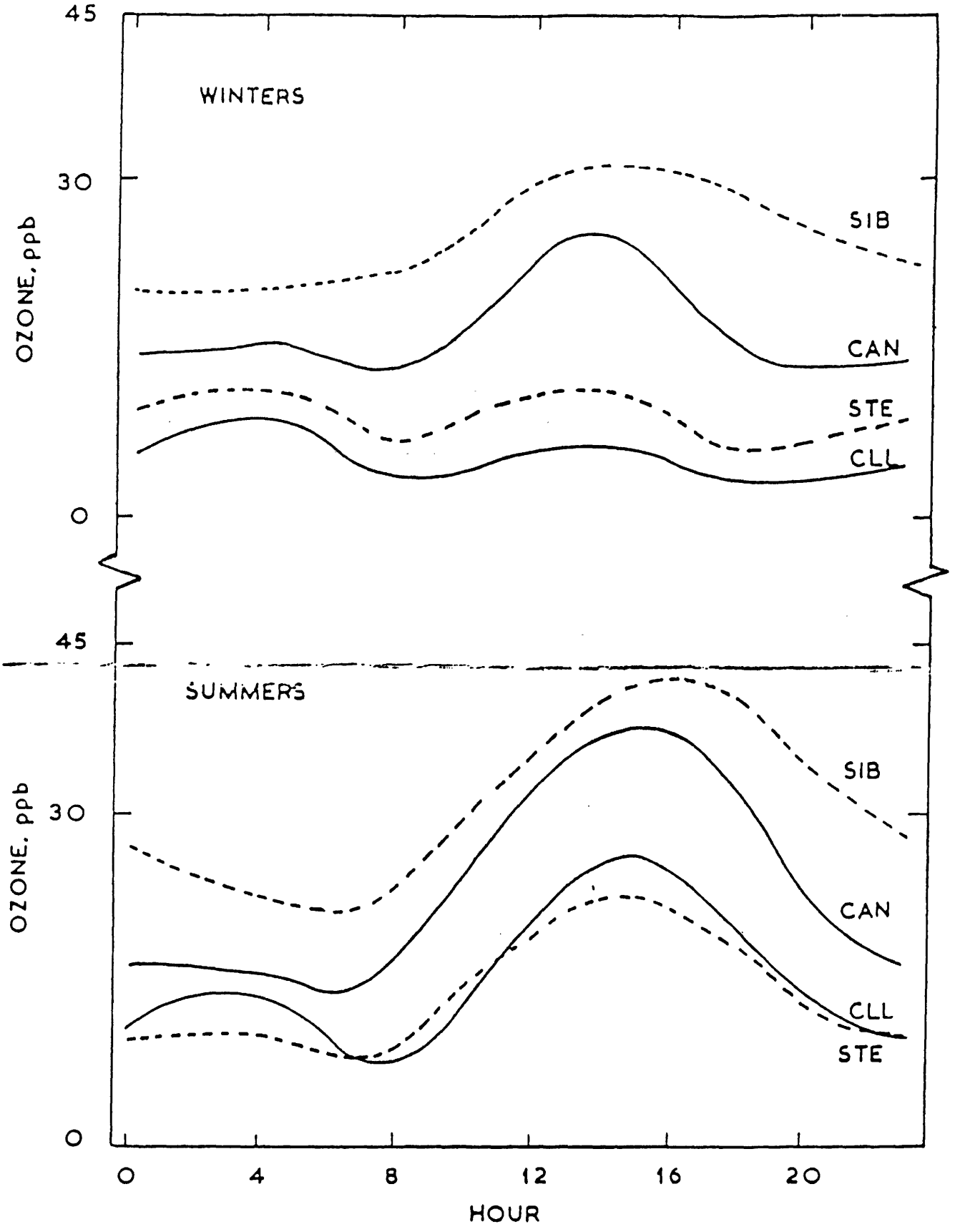

Figure III.4.5.1.-1: Diurnal patterns of O_3 at rural and urban sites

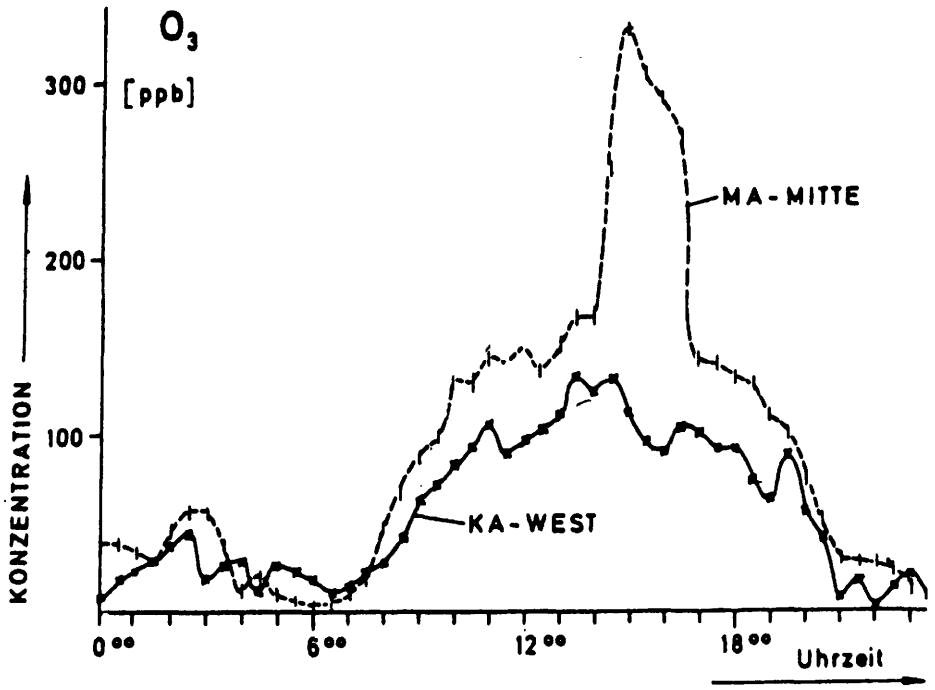

Figure III.4.5.1.-2: Diurnal variations of O_3 at two sites in Mannheim (Germany)

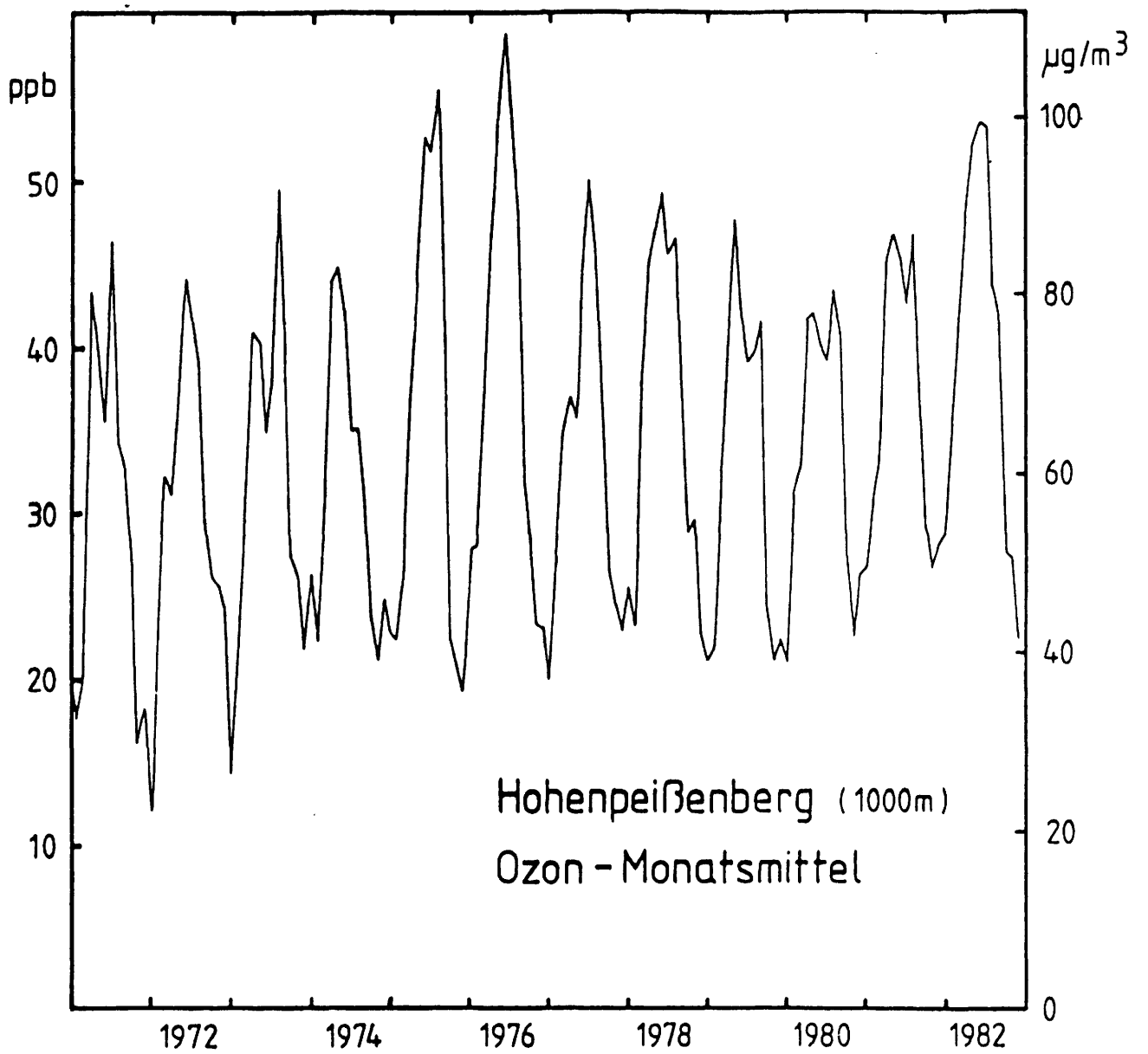

Figure III.4.5.2.-1: Seasonal variations in O_3-concentration at Hohenpeissenberg, F. R. Germany

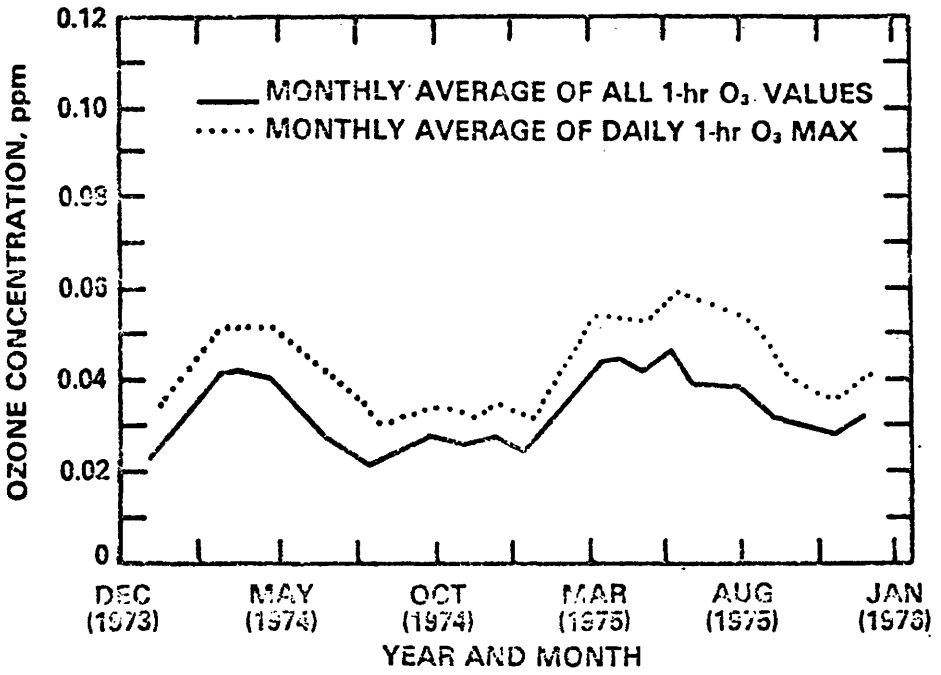

Figure III.4.5.2.-2: Long-term monthly ozone variations at Mauna Loa, Hawaii

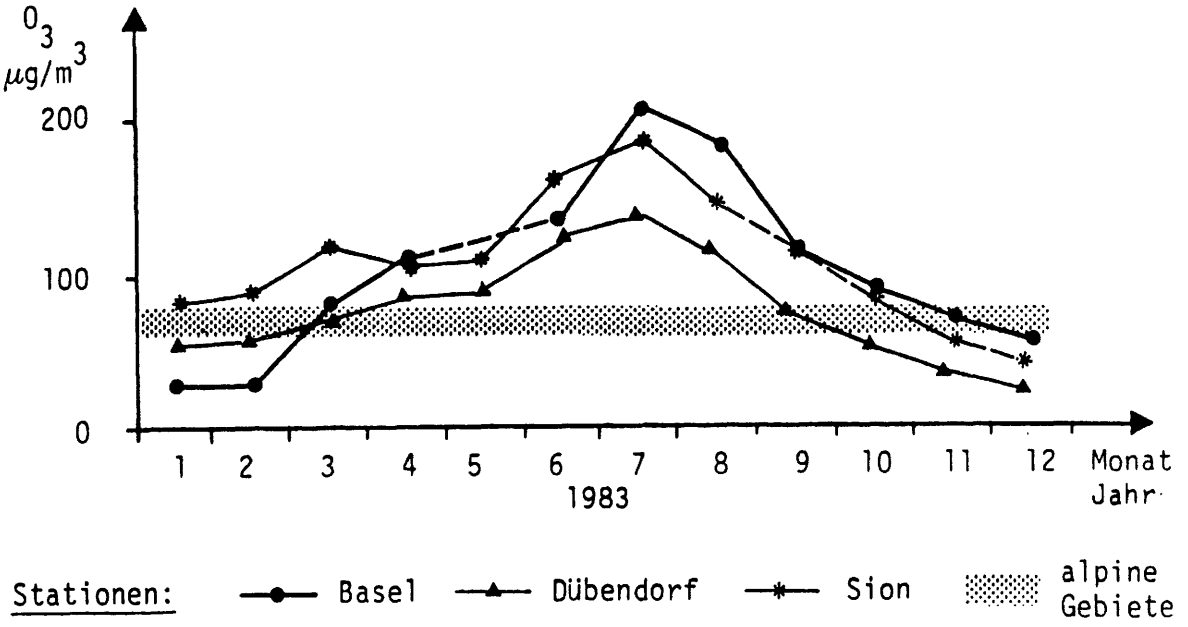

Figure III.4.5.2.-3: Seasonal variations in O_3-concentrations at three urban sites in Switzerland (monthly 95 percentiles)

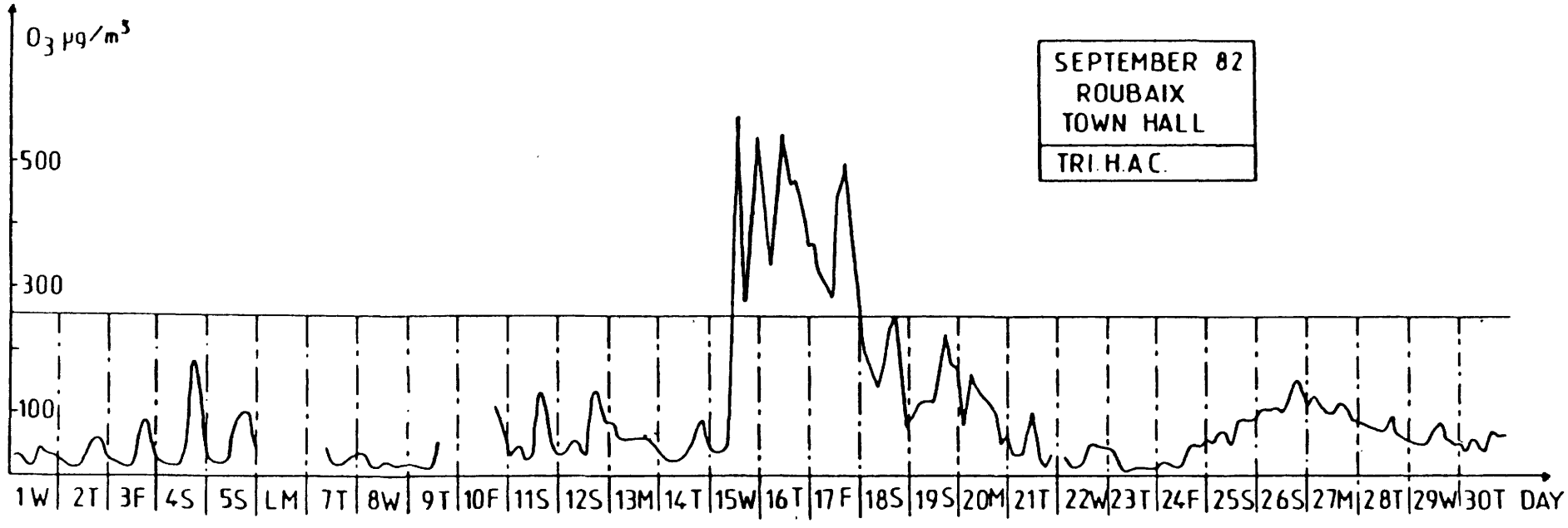

Figure III.4.5.3.-1: O_3-episode at Roubaix (France) in September 82

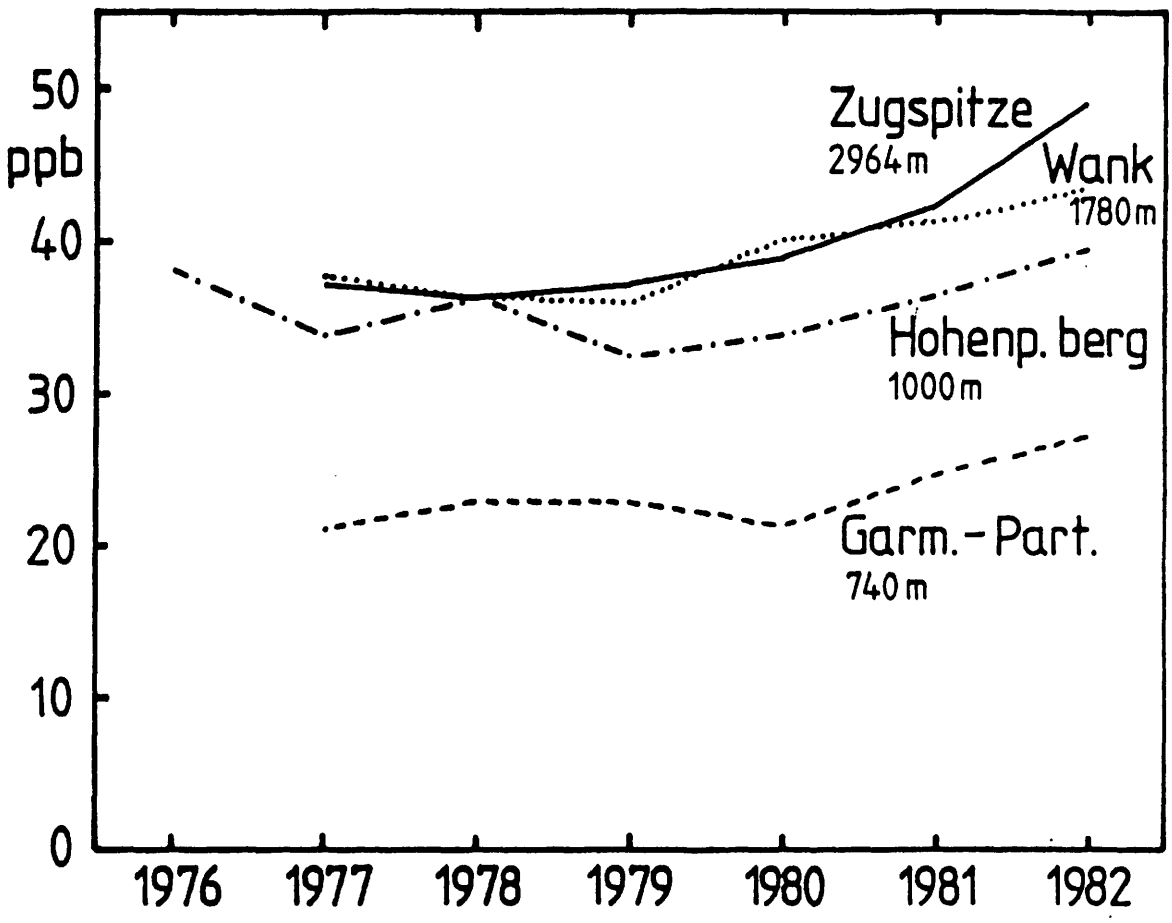

Figure III.4.6.2.-1: Trends in annual average O_3-concentration at 4 rural sites in F. R. Germany

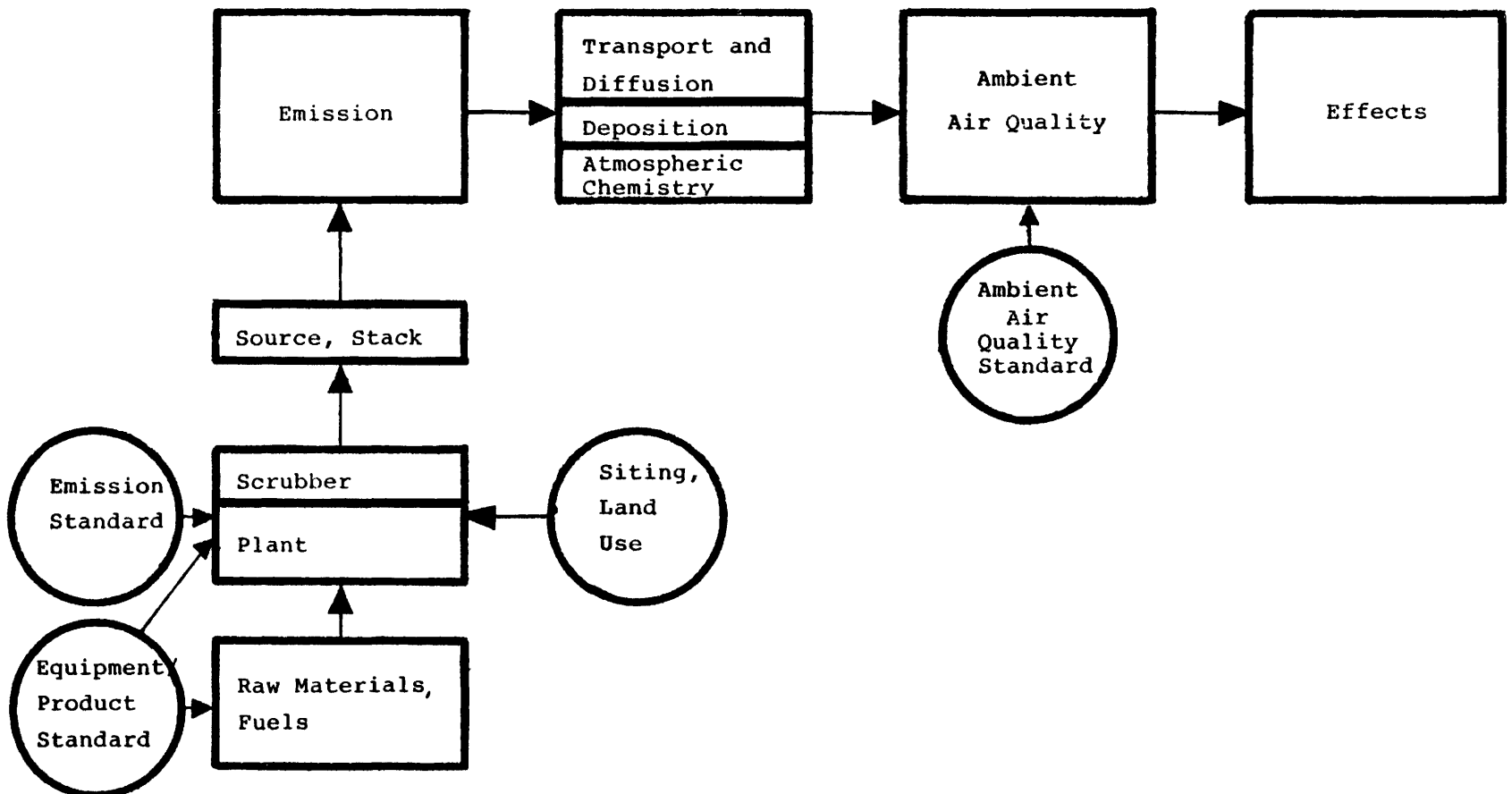

Figure IV.2.2.-1

Air Quality Management System applying Emission Standards, Equipment Standards, Product Standards, Ambient Air Quality Standards and other suitable Measures

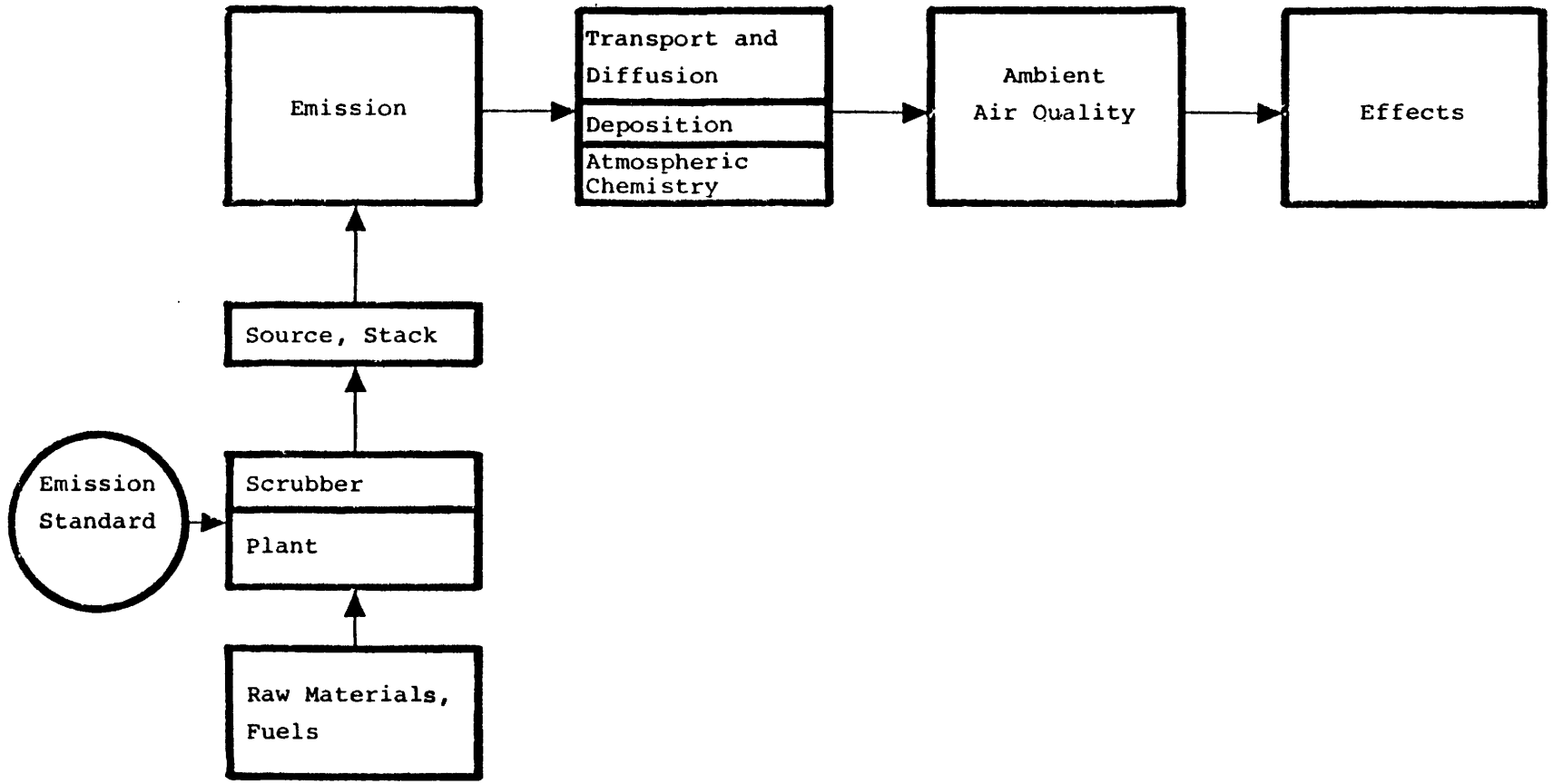

Figure IV.2.2.-2 Air Quality Management System applying Emission Standards

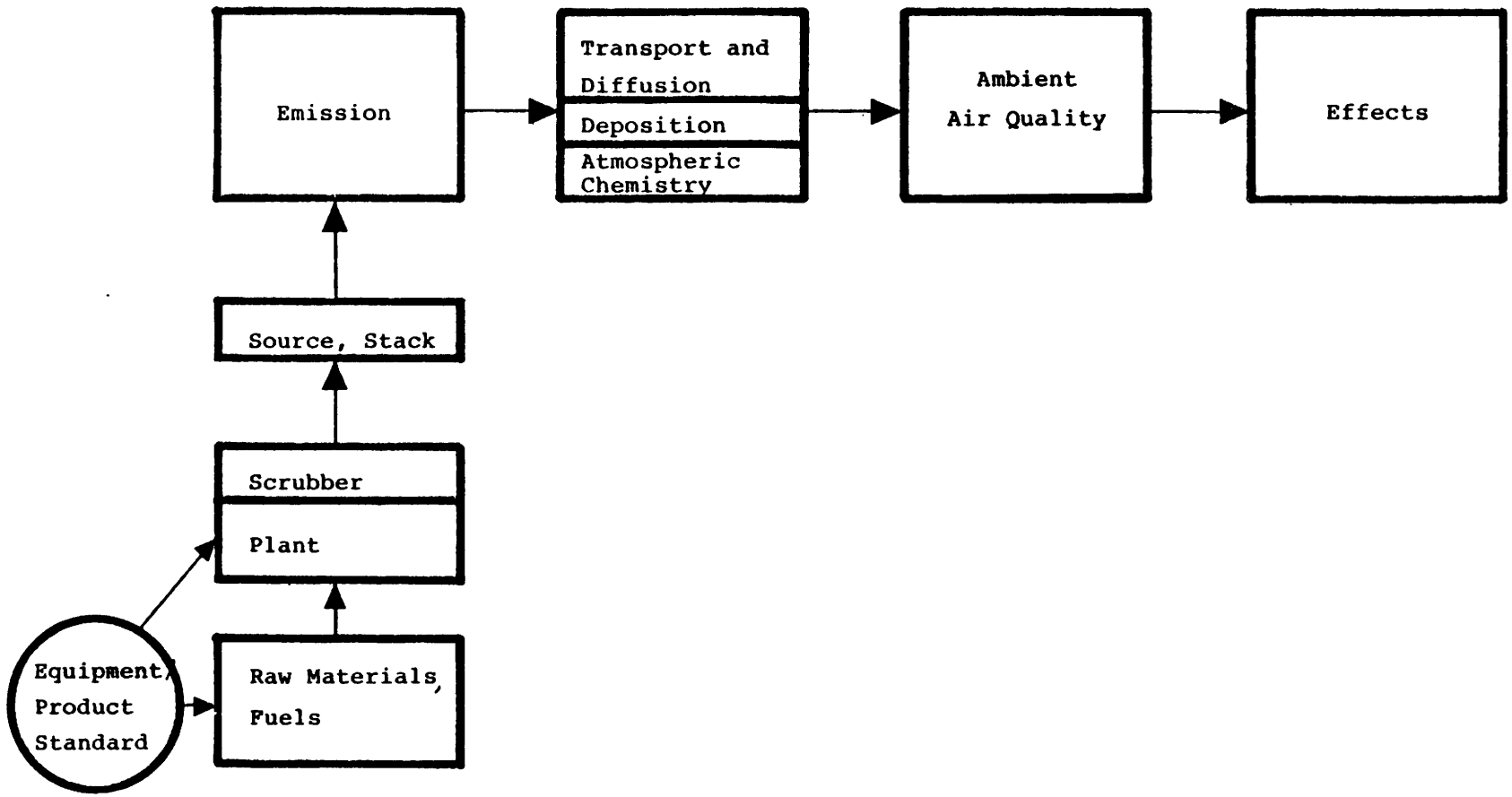

Figure IV.2.2.-3 Air Quality Management System applying Equipment Standards and Product Standards

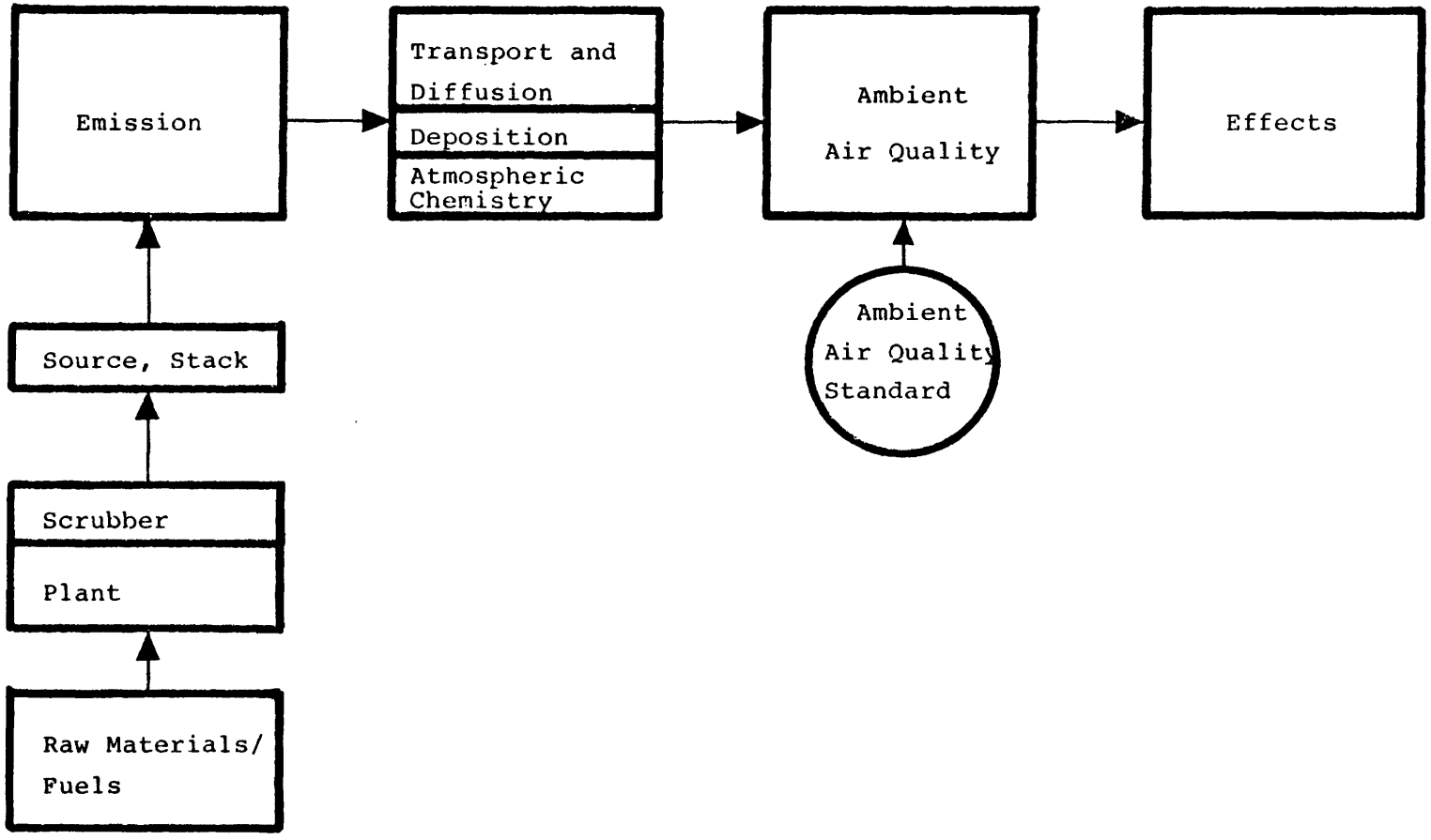

Figure IV.2.2.-4 Air Quality Management System applying Ambient Air Quality Standards

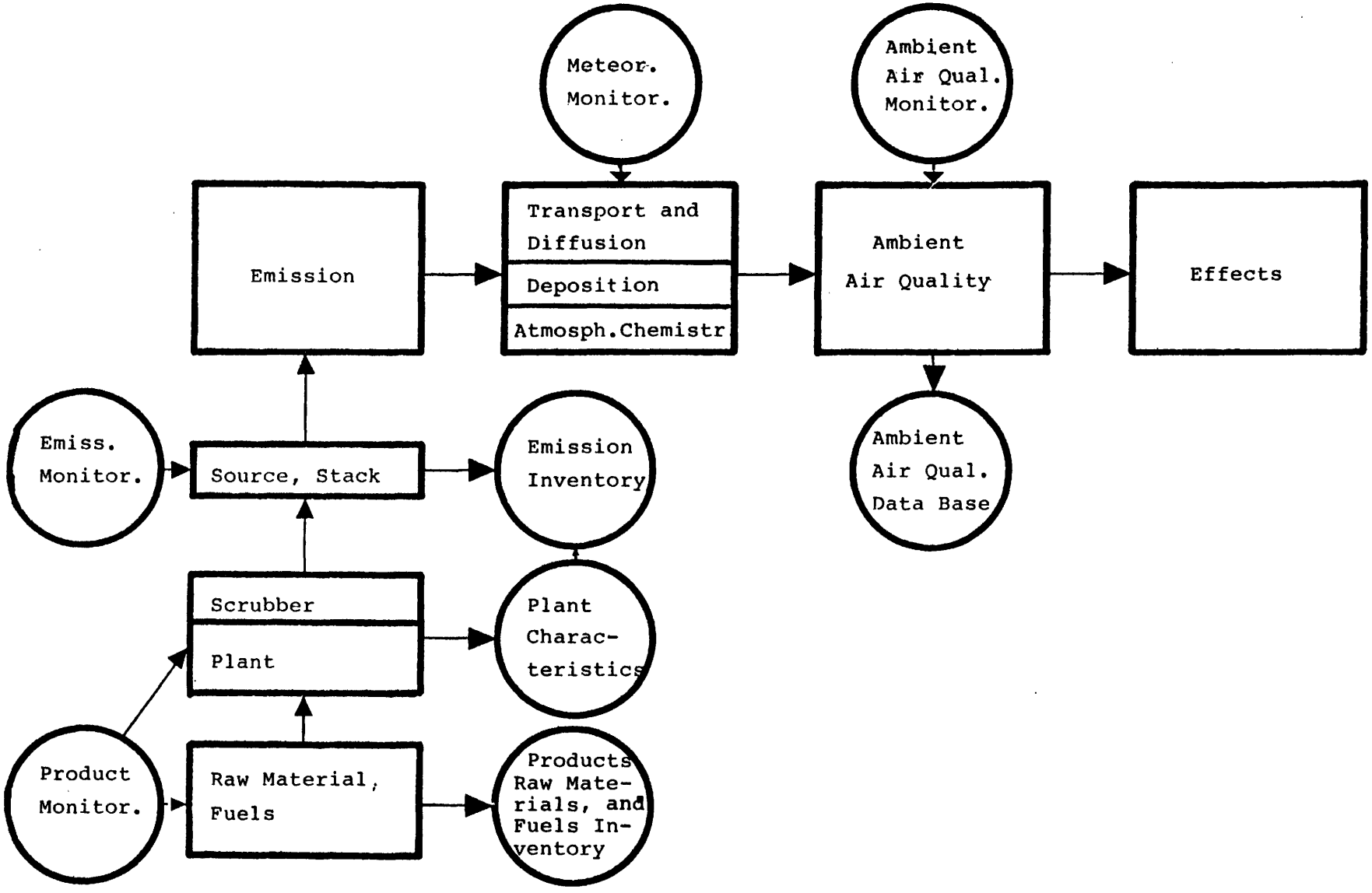

Figure IV.3.5.-1 Air Quality Management System

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The aim of the study was:

(i) compiling information on limit and guide values set out in industrialized countries and to identify those which are actually combined ones;

(ii) reviewing the relevant literature on combined effects of the pollutants NO_2, SO_2 and O_3 and particulate matter on human health and the environment;

(iii) deriving, if possible, non-adverse effect levels for combinations of these pollutants;

(iv) comparing these combined non-adverse effect levels with values derived for the effects of single pollutants;

(v) comparing the combined non-adverse effect levels with pollutant concentrations measured in ambient air;

(vi) advising the Commission in which way the findings of this study, if necessary, could be incorporated into the actual air pollution abatement policy.

With regard to point (i), the authors identified only very few cases in which combined limit or guide values are set out in industrialized countries.

From the comprehensive review of literature on combined effects it could be concluded that human exposure studies have not consistently demonstrated any enhancement of respiratory effects for combined exposures of O_3, NO_2 and CO.

The authors noted that the combined non-adverse effect levels for SO_2 and black smoke are exceeded in several urban areas but not in rural areas, for SO_2 and NO_2 these levels are most likely exceeded in some urban areas but not in rural areas and for SO_2, NO_2 and O_3 these levels are most likely exceeded in urban areas but not in rural areas.

The authors mentioned that to respect combined non-adverse effect levels, the only appropriate policy is:

— to reduce the relevant emissions in urban areas down to those levels identified as non-adverse effect levels;

— to introduce a long-term policy of an overall reduction of emissions from all sources important for long-range transport of pollutants.

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